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Review Article

Colloidal electrochromic photonic crystals: Insights into materials, mechanism, and performance enhancement

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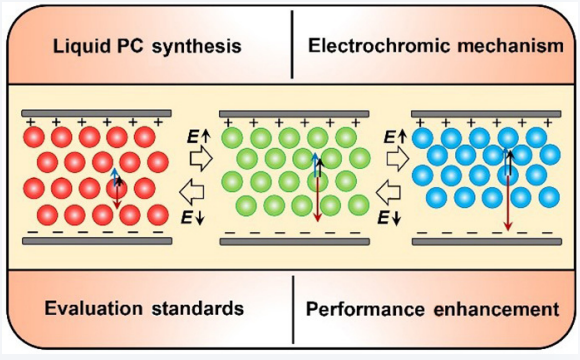
ABSTRACT: Colloidal electrochromic photonic crystals (EPCs) are optical material which changes their photonic band gap and structural colors under electric field due to the electrophoresis of colloidal particles. They have attracted great interest in recent years because of their great potential in color displays, smart windows, adaptive camouflage, and various functional devices. This review aims to introduce the fundamentals and recent progress of colloidal EPCs, which include the chemical composition and synthesis, the physical characteristics, the electrochromic mechanism, and the evaluation standards. Meanwhile, this review also summarizes the effective approaches to enhance the EPC performances from the perspective of colloidal particles, dispersant, electrode, and driving waveforms. Challenges and perspectives are finally provided for the future development of colloidal EPC materials.



KEYWORDS: electrochromic photonic crystal (EPC), colloidal particle, dispersant, driving waveform, electrophoresis

1 Introduction

Electrochromic photonic crystal (EPC) is an artificial metamaterial that changes its photonic band gap (PBG) structure and structural colors with the applied electric field. The PBG change can be a change in lattice constant, refractive index, or the degree of ordered structure. EPCs have attracted great interest in recent years because of their great potential in low-consumption displays [1–4], smart windows [5, 6], adaptive camouflage [7, 8], and functional optical devices [9, 10]. Therefore, great enthusiasm has been paid to the development of new EPC material systems and functional EPC devices.

There are four types of EPCs in public reports, including the electrochemical EPC [1, 11–16], electrically responsive liquid crystals [17–21], electroactive photonic crystal (PC) polymers [22–28], and the colloidal EPC [29–33]. The electrochemical EPC is a general term for all EPC systems triggered by electrochemical reactions. It can be a PC gel composed of SiO_2 colloidal crystals and a crosslinked metallopolymer network of polyferrocenylsilane (PFS) encapsulated in a cell with electrolyte [12], which changes PC structure (color) due to electrochemically-driven charge delocalization/reposition along the polymer chain and thereby the swelling/deswelling of the PFS polymer matrix. It can also be a (hollow-SiO_2)/WO_3 PC encapsulated in the cell with an aqueous solution of $LiClO_4$ [1], which changes color due to the change of refractive index of WO_3 at different voltages.

The electrically responsive liquid crystal (ERLC) is usually a mixture of cholesterics and chiral additives, which possesses a heliconical structure with a specific pitch length (P) and oblique angle (θ) under a proper voltage range. During the electric tuning, both P and θ decrease with the increase of the applied voltage, leading to the reflection blue shift and the corresponding color change. It should be noted that the reflection wavelength change can be as large as 1000 nm [17], which makes the color fully tunable from the ultraviolet to visible and infrared regions.

The electroactive PC polymer is a PC gel with opaline or lamellar structure, which deforms its lattice structure and changes color under electric field. Generally, it is also a mechanochromic PC with a changeable color under stretching, compression, or bending. During the electrical tuning, the deformation of such PC gel can be directly caused by the Coulombic attraction exerted on the polymer network, such as the poly(HEMA-co-MAPTA-PF6) inverse opal

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gel developed by Ueno and Watanabe et al. [22]. It can also be realized by the swelling/deswelling of the PC gel due to the motion of ions under electric field, such as the electrochromic EPC composed of ionic liquid and poly(styrene-block-quaternized-2-vinylpyridine) reported by Park et al. [25].

The colloidal EPC is intrinsically a special electrically responsive liquid photonic crystal (LPC), which is composed of monodisperse particles and a dielectric dispersant. During electric tuning, the LPC structure is formed, disassembled, or changed due to the electrophoresis of particles, which thereby changes the reflection signal and the structural colors.

Among these four EPCs, the colloidal EPC seems to be one of the most potential material, although its color tuning range is less than that of ERLC. First of all, the colloidal EPC has antiglare and brilliant structural colors based on their three-dimensional and highly ordered particle arrangement. The color saturation can be further improved when particles and dispersants with a high refractive index are used. Secondly, it has sensitively tunable colors due to the low direct-current (DC) driving voltage and the broad tunability of the reflection wavelength across the whole visible range. Thirdly, the structural color of the colloidal EPC can be reversibly changed by different voltages, and it can also be maintained by a constant voltage because no electrochemical processes are involved. Fourthly, it has a relatively fast response to the applied voltages, as a very short distance of colloidal electrophoresis causes a significant lattice deformation of LPC and thereby the color changes. Last but not least, the colloidal EPC has good compatibility with the commercialized electrophoretic display techniques, which also use the electrophoresis of white, black, or color particles to realize the color display and switching.

Since the first report of colloidal EPC by Korean researchers [29, 30], great efforts have been made to investigate the recipe of LPC, the electric cell, and the driving waveforms for EPC systems so that one can improve its performance in the aspects of color intensity, tuning range, reversibility, and stability. However, there still lacks a systematic review that organizes all related works from different perspectives, such as synthesis, mechanism, performance evaluation, and improvement.

The purpose of this review aims at answering the above question and introducing the recent progress of colloidal EPCs. The article starts with the structure, the chemical composition, the characteristics, and the synthesis of the LPCs with electrochromic response. Through the analysis of the forces applied to particles and the structure change of LPC in a capacitive cell, three electrochromic mechanisms, including lattice compression and expansion, colloidal concentration and redispersion, and lattice collapse and recovery, are proposed to explain the physical nature of the color change. To meet the requirements for the material comparison and development, a set of quantitative standards for the EPC system is then proposed to comprehensively evaluate its performance in various aspects. On the basis of these evaluations, detailed approaches to enhance the EPC performances are discussed and summarized from the perspective of colloidal particles, dispersant, electrode, and driving waveforms. Finally, we identify the current challenges for the EPC system and envision its potential applications in the future.

2 Liquid photonic crystals for EPC systems

A colloidal EPC system is composed of an electrically responsive LPC encapsulated between two parallel electrodes. A small amount of additive may be introduced to the LPC when a specific function is required. Once a DC power source is connected to the EPC system, it changes colors along with the tuning of working voltages. As the major component, the electrically responsive LPCs will be explored to pave the way for the latter discussion.

2.1 Structure and composition of LPCs

Recent investigations of colloidal assembly have led to the discovery of a special form of self-assembly structure, which is called LPCs [34–39]. The volume fraction of particles in the colloidal system is about 10%–30%, which is significantly higher than general colloidal solution but lower than that of close-packed colloidal crystals. Superficially, LPC behaves like a liquid with vivid structural color similar to that of the solid-state colloidal crystals. Intrinsically, it is a complicated colloidal dispersion, which is composed of solvent wrapped crystalline colloidal array and accompanied by a saturated colloidal solution. Coexistence of brightly iridescent crystalline and colorless domains in the optical microscope images supports the above statement. It is also indirectly confirmed by the long-range ordered colloidal assemblies adjacent to amorphous packing in scanning electron microscopy (SEM) images of a fully cured LPC composed of a curable dispersant, considering that the rapid curing process in several seconds doesn't change the particle arrangement. Since these two parts always coexist as an equilibrium in the system, LPC can be considered as an intermediate structure between a Brownian motional colloidal solution and an ordered colloidal crystal.

LPC composed of "colloidal particles" and a "liquid phase" possesses better flexibility in composition design and bandgap tuning compared to the solid-state PCs. On the one hand, the structural color of LPC can be tuned across the visible spectrum simply by changing the particle diameter or particle volume fraction. On the other hand, the liquid phase can be formulated at will in the aspects of polarity, viscosity, refractive index, and additives, to satisfy the requirements of different application scenarios. Such flexibility renders the LPC great potentials in various applications including colorimetric sensing [40–42], low consumption display [43, 44], smart window [45], photonic printing [46, 47], thermochromic material [48, 49], structural colored coating and fabrics [35, 50, 51], and anti-counterfeiting labels [52].

2.2 Characteristics compatible with electrochromism

Compared to the traditional solid-state PCs and gel-like PCs, the liquid PCs have unique physicochemical properties [40], which are confirmed by experimental results in Figs. 1(a)–1(c). (i) PBG and structural color: Owing to the long-range ordered arrangement of particles in solution, the LPC exhibits every hallmark of a colloidal photonic crystal, including a photonic band-gap and vivid structural color. (ii) Fluidity: Since the system contains a large fraction of liquid, the energy barrier for particles to slide past one another is drastically lowered, and the colloidal solution therefore flows like a viscous liquid. (iii) Metastability and tunability: As an intermediate structure between a disordered colloidal solution and an ordered colloidal crystal, external perturbations, including shaking, shearing, or sonication, can destroy the ordered state and drive the system back into a completely disordered dispersion. Such metastability behaves as flexible tunability when the disturbance is a tiny and precisely controlled stimulus, because a tiny disturbance,

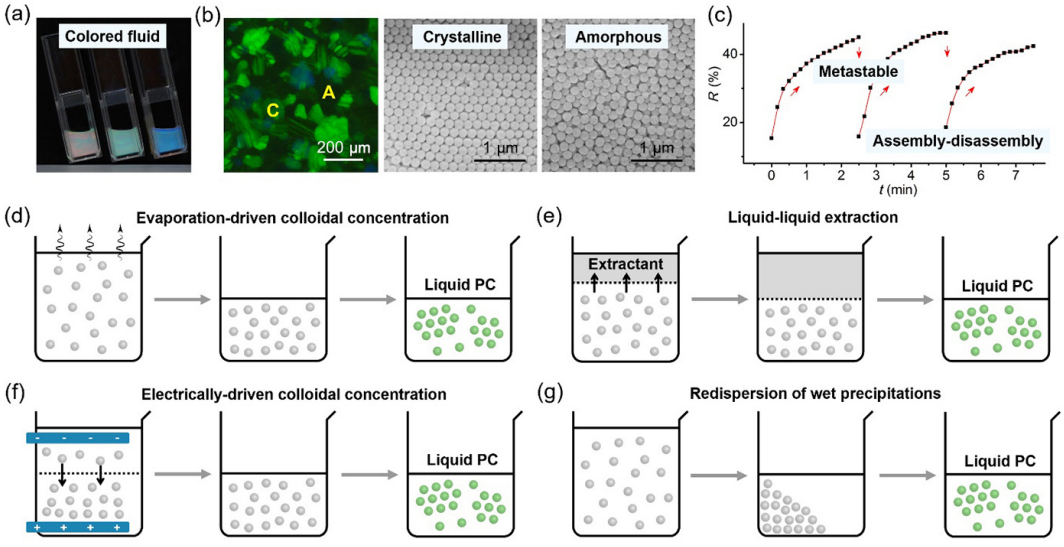

Figure 1 Characteristics and synthesis of LPC. (a) Fluidity and structural color proved by the photos of red, green, and blue LPCs. (b) Coexistence of crystalline and amorphous colloidal structures proved by the optical microscope and SEM images. (c) Metastability and reversible assembly–disassembly proved by the evolution of reflection intensity in an alternative disturbance and placement. ((d)–(g)) Synthesis of liquid PCs by evaporation-driven colloidal concentration, liquid–liquid extraction, electrically-driven colloidal concentration, and redispersion of wet precipitations. ((a)–(c)) Reproduced with permission from Ref. [40], © Wiley-VCH Verlag GmbH & Co. KGaA, Weinheim 2018.

such as a change in electrical or magnetic field strength, can change the crystal structure instead of destroying the PCs. (iv) Reversible assembly-disassembly: After the perturbation ceases and the sample is left undisturbed, colloidal particles re-nucleate and grow into the ordered colloidal crystals again. Through cycles of disturbance and placement, the colloidal particles can be assembled and disassembled repeatedly.

LPCs with specific particles and solvents are ideal materials to construct the EPC system because they are natively compatible with the electrochromic response. The aforementioned characteristics of LPC all contribute to the electrochromic applications. First of all, the LPC is a fluid with structural colors (features (i) and (ii)), which can be encapsulated in the electrophoretic cells like the commercialized liquid crystals or E-inks. Although the encapsulation destroys the ordered structure, it quickly recovers to the ordered state due to the reversible assembly and disassembly (feature (iv)). Secondly, some LPCs can respond to the electric field changes because the highly charged colloidal particles composing the LPC will migrate directionally under the electric field. Such migration can be a long-distance electrophoresis leading to a changed distribution of colloidal concentration or a short-distance movement around the lattice positions. Thirdly, the color of LPC can be effectively tuned by the applied electric field, considering the metastable and easily changed LPC structure and its good tunability under precise stimulus (feature (iii)). Through a long- or short-distance electrophoresis, the crystal lattice of LPC may expand, compress, collapse, or decompose through electric field tuning, which changes the photonic structure and thereby the structural colors. It should be noted that not all LPCs are suitable for the EPC system. The LPCs composed of hydrophobic colloidal particles usually have no electrical response.

2.3 Synthesis of LPCs

Although the concept of LPC was proposed recently, its discovery can be traced back to the 1960s [53–60], where the LPCs precipitated from a diluted colloidal solution after long-time placement. Thermodynamically, the formation of LPC is actually an entropy-driven process. During the formation of LPCs, a part of the colloidal particles precipitate to form the ordered colloidal assemblies, which is apparently an entropy decrease process. However, the rest particles achieve more space and freedom to move in the solution, which leads to an increase in entropy. Combining with these two influences, the overall process has a slight increase in entropy, so that LPCs will be spontaneously produced from the colloidal solution.

The earlier synthesis of LPCs fully relied on such spontaneous precipitation. For example, researchers noticed that an aqueous solution of polystyrene (PS) microspheres, when left undisturbed for long periods, spontaneously precipitated shiny microcrystals with structural color. Luck et al. [53] examined these microcrystals and proved that the color arises from the Bragg scattering of visible light by the ordered arrays of PS particles, which confirmed the formation of PS/H_2O LPCs. Ohno and Fukuda et al. [58] reported that the SiO_2@polymethyl methacrylate (PMMA) core–shell particles dispersed in a ternary mixture of 1,2-dichloroethane, chlorobenzene, and o-dichlorobenzene slowly precipitate to form LPCs after the solution is left quiescent for seven days. All experiments verified the entropy-driven mechanism for LPC synthesis, as colloidal particles always nucleate to form LPCs spontaneously, regardless of whether the solvent is polar or non-polar.

In the following years, the study of LPCs became very slow as it is a big challenge to synthesize LPCs in an efficient and controllable way. Several hours and even days are required for the precipitation of LPCs from the colloidal solution. We believe that the low efficiency and the poor controllability originate from the small entropy increase in synthesis. Once the entropy increase is enlarged, the synthesis shall be significantly accelerated according to thermodynamic principles. From the perspective of chemical synthesis, a highly concentrated but strictly aggregation-free colloidal solution is exactly the ideal precursor to form LPCs, because the entropy increase caused by the unassembled particles will be more remarkable in this case.

Following this idea, various new methods were recently developed to prepare LPCs from the highly concentrated but aggregation-free colloidal solution, including "evaporation-driven colloid concentration", "liquid–liquid extraction", "electrically-driven colloid concentration", and "redispersion of wet precipitations". Each method possesses its own advantages and limitations, which shall be considered when particles and solvents with specific physicochemical properties are selected to form the designed LPCs.

Yang et al. [61] reported an "evaporation-driven colloidal concentration" method, which utilized the selective evaporation of a low boiling-point (bp) good solvent to concentrate the particles to a supersaturated state in the target solvent, leading to the formation of LPCs after standing. As shown in Fig. 1(d), monodisperse colloidal particles (such as SiO_2) are dispersed in a low-bp solvent (such as ethanol or acetone) by sonication, which is mixed with a high-boiling-point target solvent with a specific volume ratio to form a homogeneous colloidal suspension. Then, the mixture is transferred to an oven at 90 °C for 2 h, during which the low-bp solvent gradually evaporates, leaving behind a supersaturated and aggregation-free colloidal suspension to produce the metastable LPCs eventually.

The key for the synthesis of LPCs, as mentioned above, is the achievement of highly concentrated but aggregation-free colloidal solution and the precise control of the particle volume fraction (f_p). Here, the f_p shall be higher than the saturated concentration of a certain colloidal particle in a specific solvent. The higher the f_p, the larger the entropy increase is achieved, so that the formation of LPCs is accelerated. Meanwhile, the f_p cannot be too high as the high viscosity severely limits the thermal motion of particles and prevents the formation of ordered particle arrangements. Generally, the particle fraction in the target solvent is designed between 0.1 and 0.35, which depends on the saturated concentration. No matter what kind of colloidal system, a particle fraction designed to be higher than 0.4 is probably unnecessary because the residual good solvent in the solvation layer of the particle makes the practical f_p lower than 0.4 according to the reflection measurement and theoretical calculation.

The remarkable advantage of this method is its high efficiency and universal applicability to a wide range of colloidal systems. It shortens the synthesis to 2 h, yet the amounts of the produced LPCs increase significantly. It is applicable to form SiO_2 LPCs in polar solvents, such as ethylene glycol (EG), propylene carbonate (PCb), N,N-dimethylformamide (DMF), and dimethyl sulfoxide (DMSO). It also works for SiO_2 LPCs in nonpolar solvents, including dichlorobenzene (DCB), aniline, amyl butyrate, and anisole, where a small amount of oleylamine is introduced to disperse the particles and enhance the surface charge of the particles. The drawback of the current method lies in the incompatibility with low-bp target solvents, so that the SiO_2/EtOH or SiO_2/H_2O LPCs can hardly be obtained.

Later, Wang et al. [62] further developed a "liquid–liquid extraction" method, which utilizes an extractant to extract part of the solvent from the colloidal solutions, concentrate the colloidal particles, and thereby obtain the LPCs after standing. As shown in Fig. 1(e), the extraction is another way to achieve a highly concentrated but aggregation-free colloidal solution.

The selection of a "suitable" extractant is the key to the above synthesis. The extractant is expected to have an intermediate polarity, which allows it to be miscible with the solvent but insoluble with the colloidal particles. Considering that the polarity of particles is usually close to that of the solvent in LPC, there is a small window for the selection of single component extract. For example, only diethyl ether (DEE) and ethyl propionate (EP) can effectively extract ethylene glycol from its solution of SiO_2 particles to produce the corresponding LPCs. Fortunately, the polarity of the two-component extractant can be precisely tuned by its ratios. Therefore, a mixture extractant composed of a non-polar solvent and a polar solvent, such as hexane/acetone and toluene/ethyl acetate, also works for the concentration of particles and fabrication of LPCs. Furthermore, some extractants have 5–6 times of extraction efficiency compared to that of DEE, so that the volume ratio of extractant and colloidal solution (V_{ex}/V_{sol}) is significantly decreased from 18 to 2.75.

Compared to the evaporation method, the extraction method requires no thermal treatment, making it especially suitable for preparing LPCs in low-bp solvents. Moreover, it is a fast, convenient, and universal method for different LPCs. It not only produces SiO_2/EG LPCs, but also succeeds with SiO_2/1,5-pentanediol, SiO_2/PCb, SiO_2/DMF, ZnO/diethylene glycol (DEG), CeO_2/DEG, and PS/EG LPCs. A notable drawback of this method is the large consumption of extractants, making it neither cost-effective nor eco-friendly. Furthermore, a small amount of residual extractant is difficult to completely remove.

Following the concentration strategy, Lu et al. [63] further developed an "electrically-driven colloid concentration" method, which utilizes the directional electrophoresis of particles to form a supersaturated colloidal solution and thereby the LPCs. As shown in Fig. 1(f), the electrophoresis is also an effective way to achieve a highly concentrated but aggregation-free colloidal solution, because the particle migration in a solution environment will not lead to permanent aggregation. In a typical synthesis, the particles are well dispersed in the solvent to form a low-concentration solution, which is transferred to a special cell with two horizontally placed electrodes. Once a vertical electric field is applied, the particles are enriched near the electrode with opposite charges to form a locally concentrated solution, which is separated by the removal of supernatant and kept standing to form LPC spontaneously.

The electrophoresis method is especially suitable for preparing LPCs composed of highly charged particles and solvents with a large dielectric constant and low viscosity, as these conditions favor fast particle migration under the electric field. It is also proven to be a universal method to prepare LPCs composed of different particles and solvents. More importantly, it could be developed into a large-scale and green synthetic process without chemical wastes through the recycling of the supernatant colloidal solution.

In addition to the above strategies, the "redispersion of wet precipitations" is reported to be a convenient and alternative way to prepare LPCs composed of specific solvents. As shown in Fig. 1(g), Yu et al. [64] utilize repeated centrifugation and redispersion by the target solvent to gradually remove the original solvent in the colloidal solution. Then, a specific amount of target solvent, depending on the composition design of LPC, is added to the "wetted" particle precipitations, which is sonicated to form the highly concentrated colloidal solution and thereafter the LPCs. Here, the retention of the wetted state for particle precipitation after the centrifugation is the key factor to form a highly concentrated but aggregation-free solution, which forces the synthesis to use repeated centrifugation to gradually replace the solvent.

Obviously, the redispersion method is a very convenient and direct way to prepare LPCs because it does not introduce any other

chemicals and requires no heating or electric field. However, this method is only applicable to the synthesis of limited LPCs, whose dispersant must have a strong capability to disperse particles. For example, it is capable of preparing SiO_2 LPCs in alcoholic and some strongly polar solvents. It is also unsuitable to prepare LPCs having critically high particle volume fraction. Furthermore, the retention of the "wetted" particle precipitations is a magic operation with great challenge when a good reproducibility is required.

3 Electrochromic mechanisms

The electric cell for colloidal EPC is mostly a capacitive cell [32], considering the insulated colloidal solution in the cell. When the external voltage is applied to the EPC cell, the positive/negative charges are injected and retained in the anode/cathode, which forms an internal electric field to store the electrical energy. Unlike a traditional capacitor, the generated electric field further drives the colloidal electrophoresis and electrochromic changes for EPC, which causes the conversion from the stored electric energy to kinetic energy of the particle and potential energy of the colloidal crystals. This is intrinsically different from the electrochemical cells, where the charges injected by the power supply go through the electrode–electrolyte interface to participate in redox reactions.

Considering the capacitive physical model for colloidal EPC, most electrochromic responses can be explained by analyzing the forces applied to the particles. The particles first experience an "electrostatic packing force" (F_{EP}) applied by the internal electric field, which is noted by a black arrow in Fig. 2. It usually drives the electrophoresis of particles towards one of the electrodes, which leads to the compression or expansion of EPC depending on the sign of the voltage.

When the voltage is not zero, there exists a "net interparticle repulsive force" between the analyzed particle and its neighboring particles because the particles with the same charges are closely arranged. Generally, these particles are arranged in a colloidal crystal lattice whose <111> direction is perpendicular to the electrode and parallel to the applied electric field. The overall interparticle repulsion from 6 particles in the same layer (F_n) equals zero due to the highly symmetric structure. However, a "net interparticle repulsion" ($F_{n-1} - F_{n+1}$) is produced by the repulsion from the 3 particles in the upper layer and the 3 particles in the bottom layer, which are noted by red and blue arrows in Fig. 2, respectively. It should be noted that the inequality of F_{n-1} and F_{n+1} originates from the non-uniform lattice spacing along the electric field direction, which can be proved by the broadening of the reflection peak along with the increase of applied voltage [31]. In most cases, the net interparticle repulsion balances with the F_{EP} to maintain a stable PC structure, and it also helps to recover the PC structure when the voltage is set to zero.

Furthermore, the viscous resistance (F_{vr}) applied to the particle also plays an important role in the electric tuning. The resistance may come from the viscous solvent or the particle itself. The F_{vr} is extraordinarily strong when the EPC lattice is highly compressed. No matter what kind of lattice change, the F_{vr} is always a resistance that needs to be overcome to achieve effective tuning.

Since the pioneering works by Korean researchers, various EPC systems have been developed, but their electrochromic mechanisms are different due to different chemical compositions, initial colloidal concentration, and the applied voltages. To explain the major mechanisms clearly, let's imagine the physical changes when a gradually enhanced voltage is applied to a colloidal system with a low particle fraction at the beginning. As shown in Fig. 2(a), in the absence of an electric field, state 1 refers to a colloidal solution with low f_{SiO_2}, in which the particles are in random Brownian motion. As the electric field enhances, state 2 corresponds to an ordered colloidal crystal with locally medium f_{SiO_2}, which is assembled due to the electrophoresis induced concentration of the particles. With the further enhancement of the electric field, state 3 indicates a colloidal crystal with a compressed lattice structure and locally high f_{SiO_2}. Its formation is also attributed to the electrophoresis, and the difference is that it's a short-distance movement around the lattice point. Eventually, an over-strong electric field leads to the collapse of the colloidal crystal into disorderly particle stacking, which is noted as state 4. The major electrochromic mechanisms are usually related to the switching between two of the above states, which will be introduced in detail as follows.

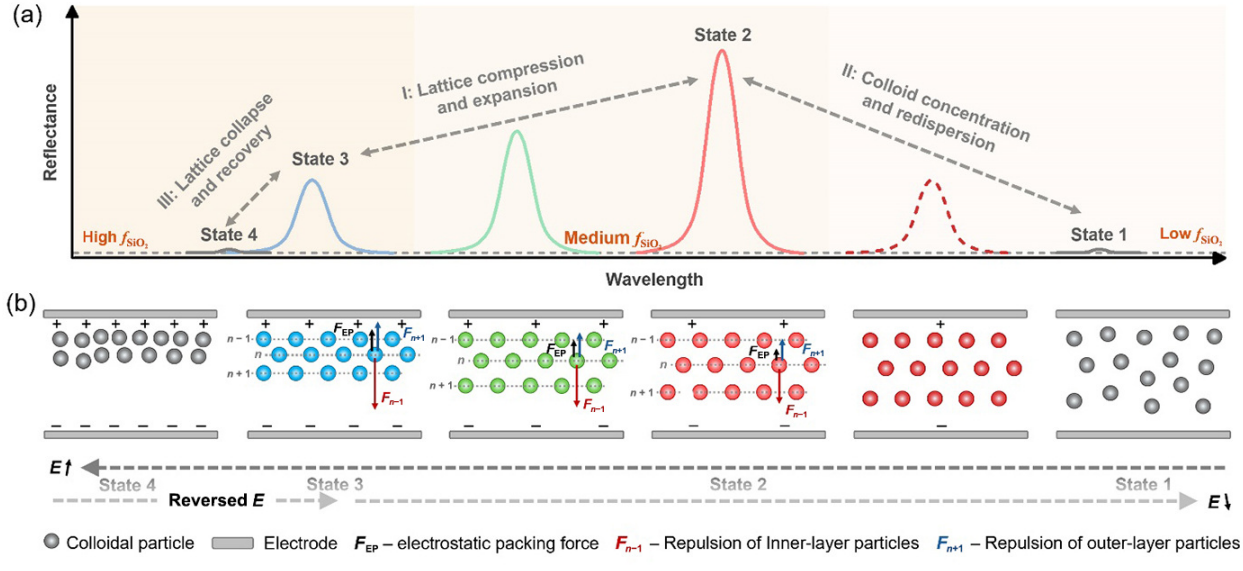

Figure 2 Three electrochromic mechanisms. The "lattice compression and expansion", "colloid concentration and redispersion", and "lattice collapse and recovery" mechanisms explained by the change of (a) reflection signal and (b) colloidal crystal structure under increasing, decreasing, or reversed voltages.

3.1 Lattice compression and expansion

The most widely adopted mechanism for the EPC system is "lattice compression and expansion", where the lattice constant changed by colloidal electrophoresis causes the change of reflection wavelength and the structural color. It corresponds to the switching between states 2 and 3 in Fig. 2(a).

For any EPC state between states 2 and 3, a structural color can be maintained at a specific voltage because of the balance of the F_{EP} and net interparticle repulsive force ($F_{n-1} - F_{n+1}$). As the applied voltage increases, the balance is broken due to the increase in F_{EP}. The EPC lattice is then compressed towards one electrode through particle electrophoresis, which leads to the reflection blue shift and corresponding color changes. Since ($F_{n-1} - F_{n+1}$) increases with the compression, it finally reaches a balance with F_{EP} so that a new color state is achieved. The reflection intensity usually decreases along with the blue shift due to the disturbance of the strong field to colloidal assembly. On the contrary, as the voltage decreases, the EPC lattice expands and the reflection redshifts because the ($F_{n-1} - F_{n+1}$) dominates the electrophoretic direction at this time. Since ($F_{n-1} - F_{n+1}$) decreases with the expansion, it eventually balances with F_{EP} to achieve the new color state. Meanwhile, the reflection intensity usually increases with the red shift due to the decreased disturbance to the colloidal assembly.

The mechanism of "lattice compression and expansion" is fully supported by the calculation of interparticle spacing through Bragg's law [65, 66]. In Eq. (1), m and λ represent the diffraction order and reflection wavelength, respectively, D is the interparticle spacing between neighboring particles, n_{eff} is the effective refractive index, and θ is the incident/reflected angle of the EPC. The effective refractive index can be calculated by Eq. (2), where n_i and f_i represent the refractive indices and volume fractions of each component of EPC, respectively. During the electric tuning, n_{eff} certainly changes with the lattice change. But it can be approximately regarded as a constant considering the closed n between particle and dispersant and maximumly 20%–30% lattice change. Therefore, the reflection wavelength change is primarily caused by the change in interparticle spacing. The calculation reveals that the interparticle spacing increases/decreases with the red shift/blue shift of the reflection peak, which well supports the electrochromic mechanism of "lattice compression and expansion".

$$m\lambda = \sqrt{\frac{8}{3}} D \sqrt{{n_{eff}}^2 - \sin^2\theta} \quad (1)$$

$${n_{eff}}^2 = \sum n_i^2 f_i \quad (2)$$

For example, the (Fe_3O_4@SiO_2)/PCb EPC system reported by Lee et al. [29] mostly follows the above electrochromic mechanism. As the voltage increases from 0 to 1.0, 2.5, and 4.0 V, the reflection wavelength gradually blueshifts and the structural color changes to red, green, and blue, respectively. In the research of Shim et al. [30], a forward voltage-induced reflection blue shift combined with a reversed voltage-induced reflection red shift also follows the above mechanism. This is probably because the EPC cell has a relatively larger thickness, so the reversed electrophoresis of part of the particles near the bottom electrode leaves adequate space for the lattice expansion near the top electrode. Such a derived mechanism is also reported by Song et al. [67] in their (Fe_3O_4@C)/acetone EPC system.

Overall, the "lattice compression and expansion" mechanism is usually suitable for an EPC system with a medium particle volume fraction (f_{SiO_2}: 10%–25%) in the initial state. The driving voltage ranges from 0 to 4 V for polar EPC, or 0 to 2 V for weak-polar EPC. The electric tuning majorly causes the change of reflection wavelength and thereby the color hue from red to blue. Such tuning is also accompanied by a decrease in color intensity at high voltages. The short-distance particle electrophoresis theoretically makes the above color change a fast process.

3.2 Colloid concentration and redispersion

The EPC system can also adopt "colloid concentration and redispersion" mechanism, where the electrophoresis drives the colloidal concentration and assembly, and the removal of electric field leads to the colloidal disassembly and redispersion. It corresponds to the switching between states 1 and 2 in Fig. 2(a).

For any EPC between these two states, there is also a balance between the F_{EP} and ($F_{n-1} - F_{n+1}$). A similar blue shift/red shift due to the voltage increase/decrease can be observed in the electric tuning. However, both of them are much weaker compared to the forces in the previous mechanism, so that the increased F_{EP} mostly facilitates colloidal assembly rather than disturbs the assembly. Therefore, the reflection intensity usually increases with the applied voltage.

For example, the TiO_2/EG EPC reported by Shim et al. [32] follows the above electrochromic mechanism. When a voltage of −3 V is applied, the positively charged TiO_2 particles are concentrated and assembled near the top indium tin oxide (ITO) electrode, showing a characteristic reflection peak at 352 nm. When the voltage is switched to +3 V, the reflection intensity gradually decreases to zero as the particle disassembles and redisperses in the solution. No reflection wavelength change is observed because the electrostatic repulsion between silane-modified TiO_2 particles is weak and the particles only assemble in a short interparticle spacing.

Overall, the "colloidal concentration and redispersion" mechanism usually works for an EPC system with a low particle volume fraction ($f_{SiO_2} < 10\%$) in the initial state. The driving voltage should be lower, considering the weak F_{EP}, but it also ranges from 0 to 4 V for practical electrical tuning, because most EPC works through this mechanism at low voltages and then through lattice compression–expansion at high voltages. The electric tuning majorly causes the change of reflection intensity, meaning that the color brightness rather than the color hue can be controlled by this mechanism. Since the particle concentration or redispersion is achieved by a long-distance electrophoresis, the electrochromic response takes a longer time than that of lattice change.

3.3 Lattice collapse and recovery

A special mechanism for the EPC system is "lattice collapse and recovery", where the collapse or recovery of the LPC structure under external electric fields causes the change of reflection intensity and the color brightness. It corresponds to the switching between states 3 and 4 in Fig. 2(a).

In this electrochromic process, an overpotential is usually applied to the LPC with ordered structures, which leads to the collapse of solvent-wrapped colloidal crystals and the quenching of reflection intensity. The "collapsed" colloidal system is so viscous (due to the high f_{SiO_2}) that it can hardly change its structure anymore, because the strong net interparticle repulsion pointing away from the electrode is neutralized by the F_{vr}. To achieve a reversible response, a reverse voltage is then applied to pull the particles away from the

disordered stacking and recover the ordered LPC and thereby the reflection intensity.

For example, the SiO_2/(PCb-PEG-EG) EPC (PEG = polyethylene glycol) developed by Fu et al. [43] follows the above electrochromic mechanism. In a typical electric tuning, a forward voltage of 4.5 V immediately changes the colored EPC with ordered structures into a colorless state with a collapsed structure. On the other hand, a reverse voltage of −2.5 V can collaborate with the net interparticle repulsion to restore the ordered PC structure, so that the colorless state is switched back to the colored one. It is worthy to be mentioned that the hardly-changed collapse structure is strengthened by the introduction of a highly viscous component, PEG, which renders the EPC a second colorless stable state (at 0 V) in addition to the general stable state with structural color. Based on the bistable and switchable EPCs with RGB colors, a low-power electrophoretic color display is developed to achieve better color saturation compared to the color-filter E-ink display.

Overall, the "lattice collapse and recovery" mechanism works for an EPC system with a relatively high particle volume fraction (f_{SiO_2} > 20%). There are very few reports about the EPC working through this mechanism. A driving voltage as high as 4.5 V is required to destroy the ordered structure, and a reverse voltage must be applied to recover the PC structure. The electric tuning also controls the color brightness rather than the hue, and a bistable response can be achieved considering the self-maintained collapse structure. It should be admitted that high voltage brings potential concern to the reversible response, and the recovery from the collapsed structure is a much slower process compared to the electrochromic changes of the previous two mechanisms.

In summary, there are no strict boundaries between the applicable scopes of the three mechanisms, because the reflection wavelength and intensity continuously change along with the increase of external voltages. However, approximate boundaries can be determined when people understand the major structural change of EPC within the applicable scope of each mechanism. For the mechanism of colloidal concentration and redispersion, the particle enrichment or dispersion induced colloidal assembly or disassembly, accompanied by the increase or decrease of reflection intensity, is the most characteristic physical change. For the mechanism of lattice compression and expansion, the major physical change is a short-distance particle electrophoresis-induced lattice compression or expansion, accompanied by a decrease or increase in reflection wavelength. For the mechanism of lattice collapse and recovery, the destruction of ordered particle arrangement under a strong electric field and the recovery of the ordered structure, accompanied by the turn-off/-on of reflection intensity, is the intrinsic physical change.

It should be noted that Fig. 2 is just an imaginary physical model that unifies all three mechanisms in the same material system for easy understanding and comparison. In fact, none of the reported EPC material covers all the mechanisms. However, for a practical colloidal EPC system, there could be two coexisting mechanisms that dominate the electrochromic process in different voltage ranges. In other words, the working mechanism of a colloidal EPC system transitions from one to another under specific conditions. For example, Fu et al. developed a bistable SiO_2/(PCb-PEG-EG) EPC for low consumption color display, where the reversible switching between colored and colorless states is actually realized by two coupled electrochromic processes dominated by different mechanisms. With the application of a strong voltage of 4.5 V, the EPC changes from green to blue in 2 s following the lattice compression–expansion mechanism, and it further changes to a colorless state at 5 s following the lattice collapse–recovery mechanism. Therefore, the exact mechanism for a specific EPC system should be proposed with careful consideration of the dominant physical model at different stages.

4 Evaluation of EPC performances

With the development of EPC materials, there is great demand to establish a broadly accepted evaluation standard to evaluate their performances in various aspects. The standardized assessment makes it possible to compare the materials and EPC systems reported by different research groups in the world. More importantly, it provides a powerful tool to optimize the materials towards balanced superior performances, which facilitates the conversion of laboratory innovations into industrial applications. As shown in Fig. 3(a) and Table 1, a set of quantitative standards were proposed recently [44], which includes tuning range ($\Delta\lambda$), maximum voltage (E_{max}), response sensitivity ($\Delta\lambda/E_{max}$), power consumption (P), response speed ($1/t_{sw}$), color intensity (R_{med}), reversibility (N), and stability (t_{mnt}). In this section, their definition, measurement, and influence upon practical usage will be discussed.

4.1 Tuning range

The $\Delta\lambda$ (nm) for the EPC system refers to the maximum variation in the reflection wavelength of EPC that can be achieved by tuning the external voltage. As shown in Fig. 3(b), the $\Delta\lambda$ is usually determined by the λ_0 of EPC in the initial state without the electric field and the λ_{max} of EPC in its maximum lattice compression state under a strong electric field. Here, the maximum lattice compression refers to a state where further enhancement of the electric field leads to the collapse of the ordered structure. In some EPC systems, the λ_0 corresponds to the reflection wavelength of an EPC *in-situ* formed by the gathering and assembly of dispersive particles under a weak field.

Since the structural color of PC materials is determined by their reflection signals, the $\Delta\lambda$ intuitively reflects the capability of color adjustment for the EPC system. Generally, $\Delta\lambda$ around 150 nm [29, 33] is the fundamental requirement for full color display, considering that the blue and red colors are achieved by reflection at 470 and 620 nm, respectively. However, a wide $\Delta\lambda$ around 200 nm [68, 69] is more desired in practical applications as it significantly decreases the chances for EPC working at high voltages.

An effective way to broaden $\Delta\lambda$ is the construction of an EPC with low particle concentration and large interparticle spacing, which leaves more room for lattice compression and color change during colloidal electrophoresis. However, just as mentioned in the synthesis section, the EPC is commonly hard to form under low particle concentration. Therefore, solvents with low critical assembly concentration (such as PCb and aniline) for a specific particle (such as SiO_2) can be selected to prepare the required EPC with a wide $\Delta\lambda$.

4.2 Maximum voltage

The E_{max} (V) is defined as the external voltage applied to the EPC, which leads to a maximum lattice compression accompanied by a minimal reflection wavelength of the EPC. As shown in Fig. 3(b), it

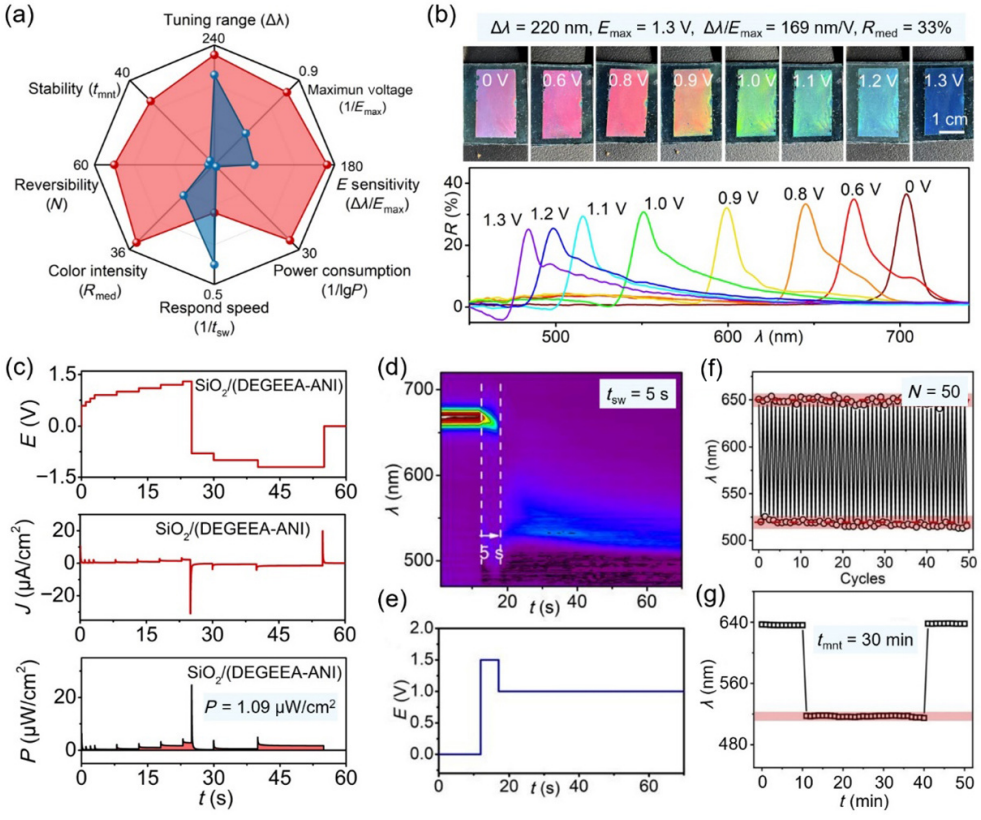

Figure 3 Evaluation of the EPC performance. (a) Comparison of the EPC performance between SiO_2/(DEGEEA-Aniline) EPC (red) and SiO_2/PCb EPC (blue). (b) Tuning range, maximum driving voltage, response sensitivity, and color intensity of SiO_2/(DEGEEA-Aniline) EPC determined from the evolution of reflection spectra under continuously increasing voltages. (c) Average power consumption determined from the time evolution of the voltage, the current density, and the instantaneous consumption. ((d) and (e)) Response time and speed determined from the dynamic reflection spectrum during the electric tuning. ((f) and (g)) Reversible cycling numbers and stable time determined by the reversible switching and maintaining of the reflection wavelength. Reproduced with permission from Ref. [44], © Elsevier B.V. 2024.

can be learned from a series of typical reflection spectra of EPC responding to the variation of external voltage. Since the colloidal assembly collapses and the reflection intensity decreases with the enhancement of the electric field, the E_{max} refers to the voltage causing the largest lattice compression but still retaining the PC characteristics.

The E_{max} is a key parameter related to many extended metrics. Combined with the $\Delta\lambda$, it can further indicate the response sensitivity of the EPC system. As the driving force of electrophoresis, it is certainly related to the response speed and power consumption. Furthermore, it also influences the reversibility and stability, as an overly high voltage leads to unwanted electrochemical reactions and particle agglomerations.

The possible way to decline the working voltage lies in the enhancement of particle surface charge and the decrease of charge screening from the dispersion media. These two strategies both enhance the Coulombic interactions between particles and electrode, so that the same lattice compression and color change can be realized under a much lower voltage. For example, a general working voltage for the polar EPC ranges from 0 to 3.5 V [29, 70]. However, for a "low-voltage EPC" with weak polar or nonpolar dispersant, the working voltage declines to the range from 0 to 1.3 V [44, 68].

4.3 Response sensitivity

The $\Delta\lambda/E_{max}$ (nm/V) of an EPC system describes the sensitive degree of the EPC structure and its reflection signals in response to the variation of external voltages. As shown in Fig. 3(b), although the sensitivity is a variable under different voltages, it is generally calculated as the $\Delta\lambda$ and the E_{max}, which reflects the average sensitivity within the whole range of working voltages.

Intrinsically, the response sensitivity reflects the ease with which an EPC system changes its color under an electric field. When the reflection wavelength change is converted into the lattice compression of an EPC, the sensitivity also indicates the average lattice change caused by a unit voltage. Under the same working voltage, a high response sensitivity means a significant change of reflection wavelength and structural color. In case of a same color change, a high sensitivity suggests a low working voltage is required to achieve that color change. Apparently, a high sensitivity is desired in practical application no matter from the consideration of power consumption or response reversibility.

The improvement of response sensitivity relies on either the increase of $\Delta\lambda$ or the decrease of working voltages. In some cases, such as the weak polar EPC system, these two requirements can be achieved simultaneously. Because an enhanced Coulombic interaction lowers the assembly concentration and working voltage at the same time.

4.4 Power consumption

The power consumption is the amount of electrical energy used by an EPC system at a specific time or over a period of time. As shown in Fig. 3(c), for an instantaneous power consumption (P_i, μW/cm²),

Table 1 Summary of chemical compositions, particle volume (or weight) fraction (f_p) of the colloidal electrochromic photonic crystals and the electrodes of EPC cell in reference works, and their electrochromic properties, including range of driving voltage (E), maximum reflection wavelength change ($\Delta\lambda_{max}$), response sensitivity ($\Delta\lambda_{max}/E_{max}$), average reflection intensity (R_{med}), power consumption (P), response time (t_{on} and t_{off}), number of color-switching cycles (N_{sw}), and the stabilized reflection change ($\Delta\lambda_{stab}$) during the stabilized period (t_{stab}). Unreported data are labeled as "—"

	Composition	f_p (%)	Electrode	E (V)	$\Delta\lambda_{max}$ (nm)	$\Delta\lambda_{max}/E_{max}$ (nm/V)	R_{med} (%)	P (μW/cm²)	t_{on} (s)	t_{off} (s)	N_{SW}	t_{stab} (min)	$\Delta\lambda_{stab}$ (nm)	Reference
1	Fe_3O_4@SiO_2/PCb	25 (wt.%)	ITO_+/ITO_-	1.0–4.0	165	41	22	—	0.05	0.17	—	600	120	[29]
2	PS/H_2O	22.1 (vol.%)	ITO_+/ITO_-	1.2–1.8	100	56	80	—	10	—	—	1–10	100	[30]
3	TiO_2/EG	6.05 (vol.%)	ITO_+/ITO_-	−3.0–3.0	—	—	—	—	—	—	—	—	—	[32]
4	ZnS@SiO_2/H_2O	> 4.8 (vol.%)	ITO_+/ITO_-	2.0–3.4	190	56	50	—	—	—	—	—	—	[31]
5	PMMA-co-PS/H_2O	—	ITO_+/ITO_-	1.5–3.5	259	74	—	—	0.2	—	—	—	—	[78]
6	PS/H_2O	—	FAS-ITO_+/ Nafion-ITO_-	2.2–3.2	150	47	—	—	0.5	8.0	200	—	—	[33]
7	PS/H_2O	—	ITO_+/DLC (or BDD-Si)$_-$	2.0–3.2	165	52	35	—	—	—	60	—	—	[79]
8	Fe_3O_4@C/PCb	18 (wt.%)	ITO_+/ITO_-	1.7–2.6	150	58	—	—	—	—	900	10	150	[76]
9	Fe_3O_4@SiO_2/PCb	12.6 (vol.%)	ITO_+/ITO_-	0.8–2.0	210	105	10	—	—	—	—	—	—	[81]
10	Fe_2O_3@SiO_2/PCb	14 (wt.%)	ITO_+/ITO_-	3.0–4.0	70	18	—	—	—	—	—	—	—	[74]
11	SiO_2/(PCb-ETPTA)	25 (vol.%)	ITO_+/ITO_-	1.0–3.3	135	41	15	—	6.0	25	—	—	—	[86]
12	CeO_2@SiO_2/PCb	17.5 (vol.%)	ITO_+/ITO_-	1.5–3.5	154	44	100	—	1.0	8.0	20	30	125	[70]
13	PtBMA/(HC-IPG-AOT)	40 (wt.%)	ITO_+/ITO_-	1.0–4.0	114	28	50	6.0	1.1	—	—	—	—	[82]
14	Fe_3O_4@SiO_2/PCb	15 (vol.%)	ITO_+/ITO_-	1.0–3.0	144	48	25	—	0.47	0.25	30	—	—	[73]
15	(PMMA-PtBMA)/ (HC-IPG-AOT)	40 (wt.%)	ITO_+/ITO_-	1.0–4.0	121	30	40	—	2.1	2.3	50	—	—	[83]
16	SiO_2/(DCB-AOT)	17.6 (vol.%)	ITO_+/ITO_-	0.5–1.7	180	106	60	—	9.0	12	100	—	—	[71]
17	SiO_2/PCb	25 (vol.%)	ITO_+/ITO_-	1.0–3.5	160	46	15	—	16	80	40	11.7	62	[80]
18	Fe_3O_4@C/PCb	25 (wt.%)	PDDA-ITO_+/ITO_-	−1.0–0.8	175	97	60	—	—	—	1000	—	—	[67]
19	SiO_2/(PCb-ETPTA -rGO)	20 (vol.%)	ITO_+/ITO_-	1.5–3.5	110	31	30	—	40	—	4	—	—	[87]
20	SiO_2/aniline	18 (vol.%)	ITO_+/ITO_-	0.5–1.4	230	164	40	—	28	15	80	30	160	[68]
21	SiO_2/(PCb-EG)	22.5 (vol.%)	Au-ITO_+/ITO_-	0.8–1.5	135	90	25	22	5.0	10	30	30	120	[72]
22	SiO_2/(PCb-EG)	22.5 (vol.%)	Au-ITO_+/ITO_-	1.5–3.0	138	46	28	—	5	90	10	—	98	[85]
23	PSMA@SiO_2/PCb	20 (vol.%)	ITO_+/ITO_-	1.0–4.0	196	49	60	—	38	154	80	—	—	[69]
24	SiO_2/(PCb-SDBS-ACNTs)	25 (vol.%)	ITO_+/ITO_-	0.5–4.0	145	36	8	—	5.0	180	10	> 2	35	[77]
25	CdS@PVP/PCb	4.5 (vol.%)	ITO_+/ITO_-	−3.5–0.0	160	46	25	—	30.0	5.0	20	3	150	[8]
26	SiO_2/(DEGEEA-aniline)	15 (vol.%)	ITO_+/ITO_-	0.6–1.3	220	169	33	1.09	9.0	11	150	30	120	[44]
27	ZnS@SiO_2/DEG	30 (vol.%)	ITO_+/ITO_-	1.5–3.5	100	29	60	—	—	—	38	> 400	45	[75]

it can be determined on the power–time (P–t) curve converted from the current density–time (j–t) and voltage–time (E–t) curves measured by the electrochemical workstation. For the average consumption (P_{avg}) during a switching or maintaining operation, it is calculated by Eq. (3) as the quotient of integrated power consumption and time for this process.

$$P = \int j(t) \cdot \mathrm{E}(t)\,\mathrm{d}t \tag{3}$$

The power consumption is another key parameter when the electrochromic response of colloidal PC is understood in the view of energy conversion. It quantifies the efficiency of the conversion from electrical energy into electrical work. Since the change of PC structure by electrophoresis and the preservation of compressed PC structure against a high electrical potential both rely on the input electrical work, the power consumption is closely related to almost all electrochromic performances, especially the response speed. Furthermore, it also demonstrates the great potential of EPC material in energy-saving display applications.

The research on methods to control the power consumption of EPC has just begun. For example, lowering the working voltage has been proven to be an effective way to reduce the power consumption by two orders of magnitude. We believe that there will be more strategies soon to accurately increase/decrease the power consumption, which helps to realize more precise tuning of structural colors.

4.5 Response speed

The $1/t_{sw}$ (s^{-1}) describes the speed of the changing of reflection signals and the structural color when a continuous or pulsive voltage is applied to the EPC system. Generally, it is defined as the reciprocal of the switching time, which is required to achieve 90% of the color change between the initial and the final states. For EPC with changeable color intensity, the switching time is related to 90% change of reflection intensity. For EPC with a changeable color hue, the switching time is connected with 90% change of reflection wavelength. As shown in Fig. 3(d), the switching time can be determined from the dynamic reflection spectrum (DRS) of the EPC measured by the ultraviolet–visible (UV–VIS) spectrometer. It can also be determined from a high-frame-rate video recording the whole color-changing process.

The response speed intrinsically reveals the speed of PC structure change caused by the particle electrophoresis. Therefore, the response speed is close to that of the commercialized E-ink display, which is another well-known electrochromic colloidal system. Generally, the structure and color change for the polar EPC system is accomplished in 1–3 s, and it can be improved to less than 1 s, which is close to the black-white E-ink display or color E-ink display with color filters. However, the switching time for the weak polar EPC system is usually as long as 10–30 s, which is close to the color E-ink display based on tri-color or tetra-color colloidal particles. A significant difference between the EPC and E-ink system is that the particle electrophoresis is short-range and long-range, respectively, which makes EPC possess faster response theoretically.

A high response speed is extraordinarily important for EPC display with the requirement of high refresh rates. The methods to accelerate the electrochromic response can be found in Eq. (4), the equation of the colloidal electrophoresis, where U is the migration speed of particles, ξ is the particle's surface potential, ε is dielectric constant of solvent, E is applied potential, K is constant, η is viscosity of solvent, and d is the distance between electrodes. One can adjust the recipes of the EPC system to achieve highly surface-charge particles, less viscous, and less screened dispersant, all of which favor the acceleration of electrochromic response. Furthermore, one can also modulate the waveform of the applied potentials to provide a precise driving force during the lattice change. For example, the "over potential modulation", "gradient potential modulation", and "reverse potential modulation" have been proven to significantly accelerate the response when the EPC lattice is compressed or expanded [71, 72].

$$U = \frac{\xi \varepsilon E}{K \pi \eta d} \tag{4}$$

4.6 Color intensity

The color intensity of the EPC system can be characterized by the intensity of its characteristic reflection peak. Since the reflection blueshifts and its intensity decreases along with the compression of the PC lattice under raised voltages, it is difficult to quantify the color intensity with a single value. As shown in Fig. 3(b), it is temporarily defined as the reflection intensity (R_{med}, %) of EPC at a medium lattice compression state. In a typical measurement, the maximum lattice compression ratio ($1 - D/D_0$) for EPC is first calculated according to the maximum wavelength change under increasing voltages, which usually equals 20%–25% in most EPCs. Then, R_{med} is determined from the reflection peak whose λ is exactly the average of the initial and final λ in the electrical tuning.

The color intensity is mainly used to characterize the color saturation and monochromaticity of the EPC during electrical tuning. A high intensity indicates a highly saturated and monochromatic structural color. The R_{med}, together with the initial intensity, also helps to evaluate the degree of "color fading" under increasing voltages. Although the previous research mostly focuses on the color change rather than the color intensities, it should be seriously considered, as an extremely weak structural color has no potential for application.

The strategy to improve the color intensity lies in the understanding of the decline of intensity under raised voltages. Simply, the decline is caused by the decrease in "periodic number of ordered structures" (N_o) due to the disturbance of the electric field to the EPC. As N_o decreases to a value that is smaller than the "maximal coherent periodic number" (N_c) of EPC to the incident light, the reflection intensity starts to decrease accordingly. Therefore, using particles with a high refractive index to construct an EPC with a small N_c is an effective way to inhibit the intensity decline. Furthermore, a low voltage with less disturbance to the ordered colloidal assembly is also applicable to maintain the color intensity.

4.7 Reversibility

The reversibility describes the capability of an EPC system to precisely and repeatedly switch its PC structure and structural color between two states under varied external voltages. When a wavelength deviation of less than 5% $\Delta\lambda$ is considered as the standard for reversible response, the reversibility can be quantitatively defined as the maximum cycling numbers (N) for reversible switching. As shown in Fig. 3(f), when a periodic driving waveform is applied to the EPC in a practical measurement, a group of reflection peaks is recorded to plot the periodic change of λ, which is used to determine N eventually. The reversibility intrinsically reveals the physical and electrochemical stability of the EPC system in repeated lattice changes. In physical aspects, the deposition of colloidal particles on electrode screens the Coulombic attraction and weakens the packing force applied on the particle towards the electrode. In chemical aspects, the electrochemical reactions due to the oxidation/reduction of the solvents or the electrode occur in the EPC system, which change the electrophoretic environment or the electrode surface structure. All the above processes influence the particle electrophoresis, the electrochromic response, and the reversibility. Therefore, the reversibility is a comprehensive indicator of the EPC, and it is also used to reveal a dominant influence in some cases.

The possible methods to improve the reversibility can be found in the analysis of the physical/chemical processes leading to the irreversible response. For example, a surface coating or modification to the electrodes may reduce the ion diffusion [33], and a low working voltage can avoid unwanted electrochemical reactions [71]. Both strategies have been proven to be effective in improving the reversibility.

4.8 Stability

The stability is defined as the capability of an EPC system to maintain its PC structure and structural color under a specific voltage. Using a similar wavelength deviation less than 5% of $\Delta\lambda$ as

the standard for maintaining PC structure, a maximum time for reflection maintenance (t_{mnt}) is used to quantitatively evaluate the stability. As shown in Fig. 3(g), when a constant voltage or a specially designed driving waveform is applied to the EPC system after a switching operation, the λ and its instantaneous deviation from the initial value after switching are continuously recorded to determine the stable time (t_{mnt}).

The stability is frequently mentioned along with the reversibility because they both indicate the retention of a highly responsive EPC system under electric tuning. The difference is that the stability reflects the reflection maintenance in a long-term electric tuning, and the reversibility indicates the reproducible reflection switching in multiple short-term tuning. They are both influenced by the unfavorable physical/chemical changes under electric tuning. Therefore, the methods for improving the reversibility are effective in improving the stability as well.

In summary, the above performance metrics from "tuning range" to "stability" provide a preliminary standard to evaluate the EPC performance in different aspects. But how to use these performance metrics for materials in different application scenarios and what about the weighting of each metric are still questions needs to be answered by more practice in the future. For instance, all these metrics are critical for EPC materials for color display. However, for a smart window with electrically tunable transmittance, $\Delta\lambda$ and $\Delta\lambda/E$ should be replaced by ΔT and $\Delta T/E$, the color intensity is non-relevant, and consumption, reversibility, and stability shall be more weighted than the response speed. For another instance, the metrics weighting of EPC materials for video streaming and dynamic posters is different, even though they are both developed for displays. The response speed is more weighted for the former, the consumption is more weighted for the latter, and the other metrics should be considered in both cases.

5 Approaches to enhance performance

The previous section introduced the quantitative standards to evaluate the performance of the EPC system. Although some guiding suggestions have been put forward to improve performance, the specific methods still need to be implemented through material design and operational optimizations. In this section, we shall discuss the detailed approaches to enhance the EPC performances from the aspects of colloidal particles, dispersant, electrode, and driving waveforms.

5.1 Colloidal particles

The colloidal particle is a key component of the EPC system because its size uniformity and surface charge influence the orderly colloidal assembly, while its refractive index and dielectric constant influence the bandgap properties and electrophoretic mobility of the particles. These influences then further determine the color intensity, color tuning range, and response speed of the EPC system.

5.1.1 Particles with high refractive index

SiO_2, PS, and PMMA are most commonly used building blocks for fabricating colloidal EPCs, because these particles can be prepared in large scale with highly uniform size through well-established sol-gel reactions or polymerization reactions. Their surface charges and hydrophobicity can also be controlled through chemical modification, which also facilitates the colloidal assembly in different dispersants. However, all these particles have relatively low refractive indices (n < 1.6), which leads to the low refractive index contrast (Δn) between particles and dispersant for the corresponding EPCs. Low contrast is unfavorable for the electrochromic response of EPC, as such a kind of EPC dramatically loses its color intensity along with the enhancement of the electric field.

To address the above issue, a series of monodisperse colloidal particles with high refractive index, such as TiO_2 [32], Fe_3O_4 [29, 73], Fe_2O_3 [74], CeO_2 [70], ZnS [31, 75], and CdS [8] colloids, have been synthesized through controlled methods along with the rapid development of nano chemistry. Core–shell colloids like Fe_3O_4@SiO_2, ZnS@SiO_2, or CeO_2@SiO_2 particles are also prepared through surface coating to construct high-performance EPC systems.

For example, Shim et al. [32] synthesized positively charged TiO_2 (n = 2.3) particles through sol-gel reaction followed by surface modification with (3-trimethoxysilylpropyl) diethylenetriamine (DETAS). The particles are used to fabricate TiO_2/ethylene glycol EPC. Influenced by the dispersant, the TiO_2/EG EPC only changes its reflection intensity under different voltages, while the reflection wavelength remains unchanged.

Because of the good compatibility of SiO_2 particles with different solvents in EPCs, core–shell particles with silica shells are prepared to construct the EPC systems. Han et al. [31] prepared monodisperse ZnS particles (n = 2.36) through the reaction between $Zn(NO_3)_2{\cdot}6H_2O$ and thioacetamide (TAA) in the presence of catalyst HNO_3 and surfactant polyvinyl pyrrolidone (PVP). The ZnS particles are further coated with a thin silica layer and grafted with 2-(4-chlorosulfonylphenyl)-ethyl-trimethoxysilane to produce negatively charged ZnS@SiO_2 particles. Benefiting from the high refractive index of the ZnS core and the strong electrostatic repulsion between particles, the ZnS@SiO_2/H_2O EPC exhibits brilliant structural color with a high reflectance of 90%. When the applied voltage is raised from 0 to 3.2 V, the reflection peak redshifts for about 200 nm, and the reflection intensity remains at a high value below 2.0 V.

With a similar consideration in particle's refractive index, Fu et al. [70] prepared CeO_2@SiO_2 particles through a polyol synthesis of CeO_2 (n = 2.4) followed by a hydrolysis of tetraethyl orthosilicate (TEOS) on the surface of CeO_2. As shown in Figs. 4(a)–4(c), when a voltage of 3.5 V is applied to the CeO_2@SiO_2/PCb EPC to trigger a reflection blue shift of 150 nm, the reflection intensity only decreases 19.4% compared to the initial intensity at 0 V. However, when the same 3.5 V is applied to the traditional SiO_2/PCb EPC, the reflection intensity tremendously decreases 83.2% compared to the initial value. According to the color intensity indicator in Section 4.6, the R_{med} for the above two EPCs is 100% and 10%, respectively. Therefore, the particles with a high refractive index indeed address the intensity decline for the traditional EPCs at high voltages. This is because the "maximal coherent periodic number" (N_c) is adequately small for EPCs with large Δn, and the decreased "periodic number of ordered structures" (N_o), even at high voltages, is still hard to be smaller than N_c, which makes the EPC resistant to color fading at high voltages. Furthermore, due to the consistency between refractive index and dielectric constant, the CeO_2@SiO_2/PCb EPC also possesses faster electrical response, reproducible on–off switching, and better stability for long-time working.

It is worth noting that the size distribution for most high-refractive-index particles is not as narrow as that of SiO_2 or polymer

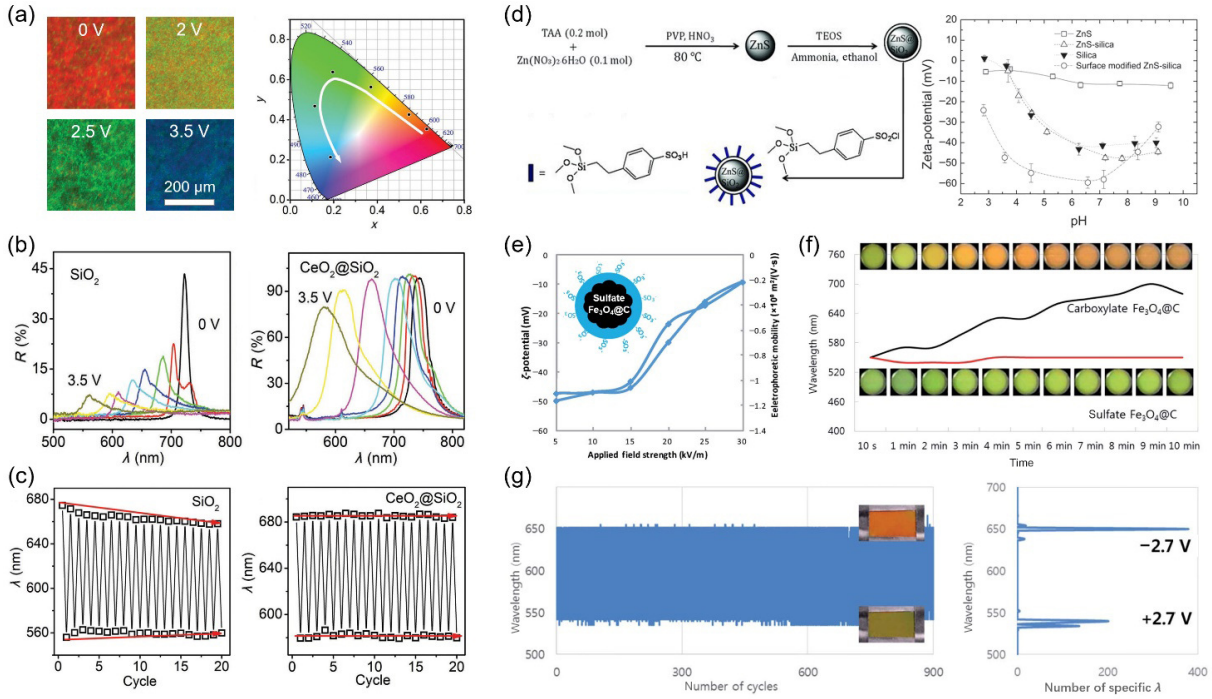

Figure 4 Performance enhancement by particles with high refractive index and high surface charge. (a) Electrochromic response of CeO_2@SiO_2/PCb EPC composed of particles with a high refractive index. ((b) and (c)) Less decline in color intensity and better reversibility for CeO_2@SiO_2/PCb EPC (high *n*) compared to SiO_2/PCb EPC. (d) Preparation of highly charged ZnS@SiO_2 particles through surface modification with 2-(4-chlorosulfonylphenyl)-ethyltrimethoxy-silane and the enhanced zeta potential due to silane modification. ((e)–(g)) Fe_3O_4@C/PCb EPC constructed by highly charged sulfate-modified Fe_3O_4@C particles, its good stability in reflection wavelength under 2.7 V for 10 min, and superior reversibility in 900 cycles of switching between 2.7 and −2.7 V. ((a)–(c)) Reproduced with permission from Ref. [70], © Wiley-VCH Verlag GmbH & Co. KGaA, Weinheim 2018. (d) Reproduced with permission from Ref. [31], © WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim 2012. ((e)–(g)) Reproduced with permission from Ref. [76], © The Royal Society of Chemistry 2016.

particles. This is usually unfavorable to the EPC system, as the broad size distribution decreases the crystallinity and thereby the color intensities. However, the decreased crystallinity also brings an unexpected advantage as these "amorphous PCs" [29, 74] with ordered structure in short-range show a structural color with angular independence, which meets the requirements of display in viewing angle.

5.1.2 *Particles with high surface charge*

It is well known that the surface charge of the colloidal particles is the key to the successful assembly of colloidal crystals. Because the orderly assembly significantly relies on the electrostatic repulsion originating from the surface charges to "place" the particles in the balanced positions in the crystal lattice.

For a dynamic EPC system, the surface charge of the particle is extraordinarily important, as it not only determines the initial assembly but also continuously influences the electrochromic response in the following electric tuning. Fortunately, the most widely used silica and polymeric particles for the EPC system usually carry a decent surface charge after their synthesis. The SiO_2 particles possess a negative charge as the surface Si–OH groups disassociate to release H^+ into the solvents, leaving behind a charged particle, while the PS or PMMA particles also have a negative charge in their aqueous solution because the persulfate initiator residuals remain at the end of the polymer chains. Although these particles already have a specific amount of charge, a stronger surface charge is always desired for better electrochromic performance. Therefore, various strategies, including copolymerization, silane coupling, and surface modification, are developed to achieve that objective.

Copolymerization is an effective method for the modification of polymer particles. In the earlier study by Shim et al. [30], the PS particles are grafted with additional sulfonate groups through copolymerization with 3-allyloxy-2-hydroxy-1-propane-sulfonic acid sodium salt. The sulfonate modification renders the PS particle a surface charge density of −6.026 μC/cm² and a zeta potential of −54.3 mV, which enables the PS/H_2O EPC to achieve a 70-nm wavelength tuning under a 1.6 V alternating current (AC) electric field with fast and stable response.

The silane coupling is an effective method for the modification of SiO_2 particles. For example, Shim et al. [32] prepared 103-nm TiO_2 particles through sol-gel reaction and modified the particles with a silane coupling agent, 3-trimethoxysilylpropyl-diethylene-triamine (DETAS). The positively charged TiO_2 particles with a typical zeta potential of +56 mV are then used to fabricate TiO_2/EG EPC with a dramatic change of reflection intensity in response to the applied electric field of +3/−3 V. As shown in Fig. 4(d), Han et al. [31] modified the ZnS@SiO_2 core–shell particles with a silane coupling agent, 2-(4-chlorosulfonylphenyl)-ethyltrimethoxy-silane, where the grafted chlorosulfonyl groups (–SO_2Cl) are hydrolyzed to produce stronger negative surface charges. Through silane modification, the zeta potential of the particle enhances from −48 to −60 mV at a pH of 7. Thanks to the high surface charge and high refractive index of the colloidal particles, the ZnS@SiO_2/H_2O EPC achieves a bright color tuning throughout the whole visible range.

In addition to the above methods, surface modification with charged chemical species is a general way for more particles. As shown in Figs. 4(e)–4(g), Lee et al. [76] modified the *in-situ* produced Fe_3O_4@C particles with sulfate groups in a solvothermal reaction with the presence of potassium persulfate. Compared to

the carboxylate-modified Fe_3O_4@C particles, these sulfate-modified particles are highly stable without deposition onto the electrode during electrophoretic operation due to inert sulfate groups. Therefore, the sulfate-Fe_3O_4@C/PCb EPC exhibits superior stability and reversibility in electrical tuning. Under a static 2.7 V electric field, the Fe_3O_4@C/PCb EPC shows no reflection wavelength change in 10 min. When voltages of 2.7 and −2.7 V are alternatively applied, the EPC shows reversible reflection wavelength and intensity for hundreds of cycles with fluctuation less than 5%.

Following the same strategy, Wang et al. [77] modified SiO_2 particles with sodium dodecyl benzene sulfonate (SDBS) and prepared a (SiO_2-acidified carbon nanotube (ACNT))/PCb EPC with electrochromic response. Here, the anion groups provided by SDBS are introduced into the slipping layer of SiO_2 particles, leading to the high surface charges for colloidal particles, while the black material ACNTs enhance the color intensity by absorbing incoherently scattered light. The high surface charge further improves the response rate and enhances stability and reversibility.

5.2 Liquid dispersant

The liquid dispersant, usually a solvent of the LPC, is another core component of the EPC system, as its polarity, refractive index, viscosity, and chemical properties significantly influence the dispersibility, the surface charge, and the electrophoretic mobility of the colloidal particles. Through these influences, it further affects almost all the properties of EPC so that screening a suitable dispersant becomes a quite effective way to improve the performance of the EPC system.

5.2.1 Polar dispersant

Polar solvent is the primary choice to construct the earlier colloidal EPC systems because a stable dispersion of particles in the system and a high surface charge for electrophoresis are the basis for electrochromic response. It is well known that the polymeric particles and silica particles are the major building blocks to construct the LPCs. These particles usually have ionic surface groups and polar surfaces, which make them only dispersible in water and polar organic solvents. Meanwhile, the charge separation processes, including the dissociation of H^+ from the Si–OH, COOH, and SO_3H groups, are much easier in the polar solvent so that the particles obtain an adequately high surface charge for electrophoresis and electrochromic changes.

Generally, water is used for polymer-particle-based EPCs. In the pioneer work, Shim et al. [30] reported a PS/H_2O EPC, which blue-shifted its reflection for 100 nm and presented corresponding color changes when a DC field was applied and gradually increased from 0 to 1.8 V. In the following research, the (PMMA-co-PS)/H_2O [78], (ZnS@SiO_2)/H_2O [31], and other PS/H_2O [33, 79] EPCs were successively developed to exhibit similar electrochromic responses.

PCb is another frequently used solvent for silica-based EPCs. PCb is a strongly polar solvent with a dielectric constant of 64, and its good stability against chemicals and electrolysis makes it an ideal electrolyte solvent in lithium-ion batteries and other electrical cells. As a nonaqueous solvent, it avoids the electrolysis of water and release of ions during the electrical tuning, which effectively improves the reversibility and stability of the EPCs. For example, Lee et al. [29] reported the (Fe_3O_4@SiO_2)/PCb EPC, which is the first of such kind. This EPC not only possessed tunable structural color under voltage from 0 to 4 V, but also showed a stable color for 10 h under 2.5 V, which suggested no redox reaction occurred in the PCb solution. Illuminated by this work, the SiO_2/PCb [80], the (Fe_3O_4@C)/PCb [76], the (α-Fe_2O_3@SiO_2)/PCb [74], the (CeO_2@SiO_2)/PCb [70], and other PCb-based EPCs [69, 81] were prepared gradually, which further confirmed the advantages of the polar EPC system.

5.2.2 Weak polar dispersant

Early EPCs primarily utilized strongly polar solvents, such as H_2O and PCb, to achieve particle dispersion and high surface charges. However, the polar solvent causes a strong shielding of the Coulombic interaction between particles and electrodes. Because the polarized solvent molecules generate an induced field (E_{ind}) with opposite direction to the external field (E), which lets the effective field (E_{eff}) applied to the particles equal to the subtraction of E_{ind} from E. Therefore, a voltage as high as 3.5–4 V is usually required to drive the particle electrophoresis to realize the largest lattice compression and largest color changes. Such high voltage inevitably triggers electrochemical reactions to change the state of LPCs and electrodes, leading to the degradation of performance eventually.

To address this issue, a series of weak-polar EPCs have been developed in recent years. Since the weak polar solvent has much weaker shielding to the Coulombic interaction, these EPCs can work at much lower voltages, showing lower power consumption and better reversibility/stability. The real challenge is the well dispersion of polar particles and the formation of highly charged particles in weak-polar solvents. Great efforts have been made to conquer the above difficulties.

As shown in Figs. 5(a) and 5(b), Ko et al. fabricated a PtBMA/(HC-IPG-AOT) EPC [82, 83] by separating the poly(*t*-butyl methacrylate) (PtBMA) particles from the aqueous reaction solution and then dispersing them in the mixture of halocarbon oil (HC), isopar G (IPG, ExxonMobil), and dioctyl sulfosuccinate sodium salt (AOT) under ultrasonication. Here, the surfactant AOT molecules form charged "water-in-oil" reverse micelles by encapsulating trace moisture in the air, which stabilizes the particles through adsorption of charged micelles. In their report, the maximum electric current of the PtBMA/(HC-IPG-AOT) EPC is measured to be ~ 1.5×10^{-6} A at 4 V, which produces an extremely low consumption of 6 μW/cm^2. However, when the PtBMA particles are dispersed in deionized (DI) water, the maximum current of the PtBMA/H_2O EPC is measured to be ~ 1.5×10^{-3} A at 4 V, which suggests a 1000-fold lower consumption for the PtBMA/(HC-IPG-AOT) EPC.

Using the same AOT, Bao et al. developed a SiO_2/(DCB-AOT) EPC [71] with a different working mechanism. They modified the SiO_2 particles with dodecyltrimethoxysilane and then dispersed the particles in the mixture of 1,2-dichlorobenzene (DCB, ε = 9.8) and AOT. As shown in Figs. 5(c)–5(g), the hydrophobic modification works with the AOT micelles to promote the particle dispersion. Meanwhile, these AOT micelles stabilize the counterions (H^+) from the particle, bring them away, and leave behind negative Si–O^- groups, which effectively promote the charge separation and produce highly charged particles in the weak-polar system. When the weak-polar SiO_2/(DCB-AOT) EPC is compared with the traditional polar SiO_2/PCb EPC, one can easily find that the working voltages for the same color change of 150 nm are 1.4 and 3.5 V, respectively. The significantly lowered voltage for SiO_2/(DCB-AOT) EPC lets the colloidal assembly be less affected by the electric field and enhances the color intensity at high voltages, which can be supported by the normalized R_{med} of 0.83 compared to that of 0.57 for SiO_2/PCb EPC. Meanwhile, the low voltage inhibits the

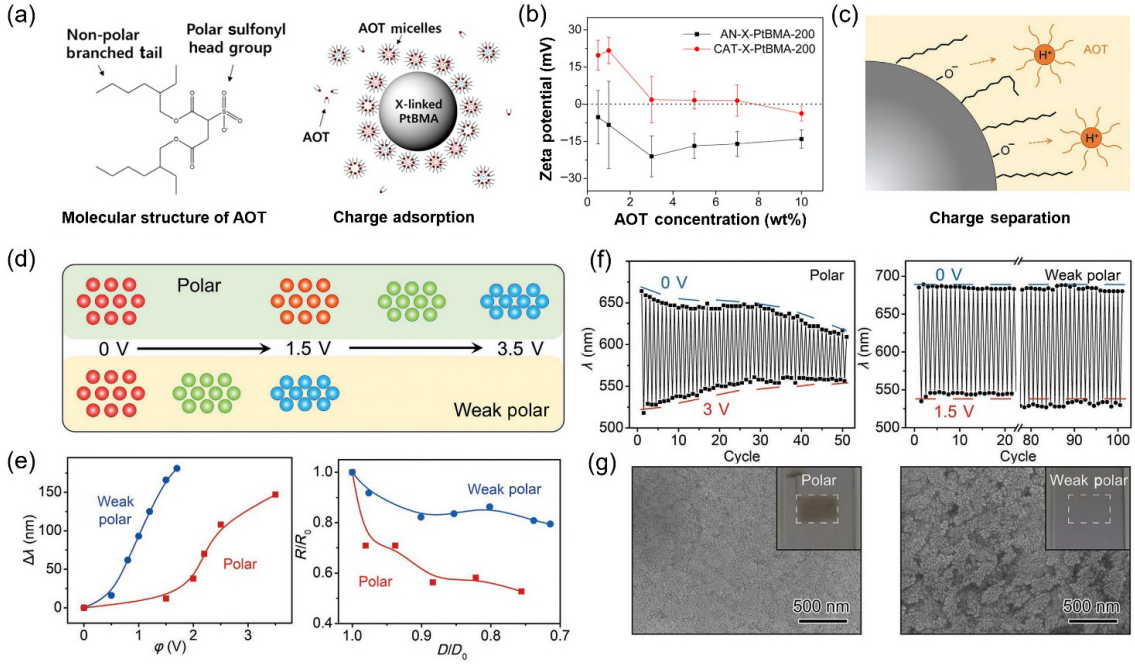

Figure 5 Performance enhancement by weak-polar dispersant. ((a) and (b)) Molecular structure of AOT and the enhancement of particle's surface charge in weak polar dispersant through the adsorption of the charged "water-in-oil" reverse micelles (AOT micelles). (c) Enhancement of the particle's charge in a weak polar dispersant due to the charge separation by AOT micelles. ((d) and (e)) Lower driving voltage, broader tuning range, and higher color intensity for weak polar EPC, supported by comparing the evolution of reflection wavelength changes with driving voltage and the evolution of reflection intensity with lattice compression for SiO_2/(DCB-AOT) weak polar EPC and SiO_2/PCb polar EPC. ((f) and (g)) Good reversibility for weak-polar EPC due to low voltage and less electrochemical reaction on the ITO electrode, as proved by the reflection wavelength switching, the electrode's photos, and SEM images for weak-polar and polar EPCs. ((a) and (b)) Reproduced with permission from Ref. [82], © American Chemical Society 2018. ((c)–(g)) Reproduced with permission from Ref. [71], © Wiley-VCH GmbH 2022.

reduction of In on the ITO electrode and renders the SiO_2/(DCB-AOT) EPC long-term reversibility during color switching, which is proved by the reversible cycling number (N) of 100 and 4 for the two EPCs.

In addition to the AOT-based EPCs, Bao et al. also developed a SiO_2/aniline EPC [68], where neither the addition of charge controlling agent (such as AOT) nor the hydrophobic modification of particles is required. As a special weak-polar solvent, aniline plays similar roles as the AOT micelles and hydrocarbon silane. Firstly, aniline molecules are adsorbed on the particle surface due to the strong interaction between Si–OH and $-NH_2$ groups, which enhances the colloidal stability. Secondly, aniline accepts a proton and brings them away from the Si–OH group, producing the negatively charged particles to self-assemble into LPCs. Compared to most EPCs, this EPC achieves a broad color change (230 nm) under a low voltage (1.4 V) due to less screening of Coulombic interaction. The low voltage brings less disturbance to the colloidal assembly, which maintains the good color intensity at high voltages. It also avoids the reduction of ITO, which gives the EPC good stability under a long-term electric field and good reversibility during the switching.

Most recently, Fu et al. reported a weak-polar SiO_2/(DEGEEA-aniline) EPCs [44] with improved performances in many aspects, including broad tuning range, low driving voltage, excellent reversibility and stability, and low power consumption. As a weak-polar dispersant, the mixture of di(ethylene glycol) ethyl ether acrylate (DEGEEA) and aniline helps the EPC achieve a color change (220 nm) under a low voltage (1.3 V). This EPC presents reversible wavelength and intensity change for 150 cycles when the applied voltage switches between 0 and 1.0/0.9/0.8 V periodically. It also shows stable reflection signals in 30 min when a constant voltage is applied. For a successive color tuning (red–blue–red) under programmed driving waveform, the EPC merely consumes 1.09 μW/cm^2, which saves 96.5% power compared to traditional SiO_2/PCb EPC (31.5 μW/cm^2). Although the weak polar dispersant promotes most of the EPC performance, the low voltage is unfavorable for the fast response, which needs to be compensated for by other methods.

5.2.3 *Alcoholic dispersant*

Alcoholic solvents are one kind of special dispersant for SiO_2-based EPCs due to the strong interaction between particles and solvents and the stable particle dispersion even at high concentration. The extraordinarily stable dispersion of particles renders the EPC very stable reflection signals, even under long-term and constant voltages. However, the stable dispersion also makes the particles orderly assemble only at high volume fractions, which shortens the initial interparticle spacing and therefore narrows the $\Delta\lambda$ under the same voltage change. In some cases, the reflection wavelength doesn't change at all, and only the change of reflection intensity is observed.

Shim et al. fabricated a TiO_2/EG EPC [32] by dispersing 6% (3-trimethoxysilylpropyl) diethylenetriamine modified TiO_2 particles in EG. This EPC reversibly shows strong/weak reflection under −3/+3 V, which corresponds to the locally concentration-induced assembly and dilution-induced disassembly of particles under opposite electric fields, considering the low particle volume fraction. No change in reflection wavelength is observed, which confirms the very short interparticle distance and poor capability in wavelength tuning.

Ye et al. also noticed the influences of alcoholic solvents in their studies [84]. For the SiO_2/PCb EPC system, the long-term stability of the reflection wavelength is significantly improved when the solvent PCb is partially replaced by polyols, among which EG has the best function. Based on such stabilization, the SiO_2/(PCb-EG) EPC is used to fabricate an electrically triggered anticounterfeiting tag [72, 85], whose invisible pattern at 0 V can be "revealed in multicolor form" or "gradually revealed" under increasing voltages.

Recently, Li et al. developed a ZnS@SiO_2/DEG EPC [75], which displayed a stable reflection at 2.5 V for over 400 min, which is much longer than the traditional SiO_2/PCb EPC. Here, DEG molecules form strong hydrogen bonds with the Si–OH groups on the ZnS@SiO_2 core–shell particles, which constructs a dense solvation layer to provide extra repulsion to maintain the lattice structure under long-term application of external fields.

5.2.4 *Functional dispersant*

Different from the dispersant aiming at the enhancement of response performance, some functional chemicals serving as the additional dispersant render the EPCs unique properties, such as structure fixing and bistable tuning. The functional dispersant includes the photo-polymerizable monomers, the viscosity enhancer, the shape memory polymer network, and possibly more substances discovered in the near future. The extra functions, combined with the electrochromic effect, naturally expand the application of EPCs in new fields.

Chen et al. designed a curable SiO_2/(PCb-ETPTA) EPC [86] with the solvent ratio of 4:1, and used it as a special ink to achieve multicolor PC printing. Here, trimethylolpropane ethoxylate triacrylate (ETPTA) is a photocurable monomer that rapidly polymerizes and fixes the electrically tuned PC structure permanently [87]. Based on this electrochromic and curable liquid ink, lithographical printing with photomask and maskless pixel printing techniques were developed to prepare multicolor and high-resolution photonic patterns.

Fu et al. developed a bistable SiO_2/(PCb-PEG-EG) EPC [43] and used it for low-power electrophoretic color displays. When the EPC is compressed to a disorderly particle stacking under high voltage, the high viscosity due to the introduction of PEG and EG balances the electrostatic repulsion and stabilizes the disordered structure. Therefore, in addition to the stable colored state from colloidal self-assembly, this EPC also has a stable colorless state at 0 V with disordered particle stacking. More importantly, these two states can also be reversibly switched by application of a short-time electrical field, which refers to 4.5 V/5 s for switching to the colorless state and −2.5 V/4 s for switching to the colored state. This bistable EPC indicates a new route to achieve low-power reflective displays with colloidal materials.

Xie et al. reported a bistable and reconfigurable Fe_3O_4@C/(pSA-co-pUDA) EPC [88], and used it as an ink-free rewritable paper for information recording. Here, the dispersant polystearyl acrylate-co-polyurethane diacrylate (pSA-co-pUDA) is an electroactive shape memory polymer, which not only helps to change the lattice structure of the embedded one-dimensional (1D) particle chain under electric field but also keeps the PC structure permanently until a thermal treatment is applied to recover the initial structure. Different from most EPCs, the pSA-co-pUDA is a solid dispersant, which opens a large space for the design and fabrication of new colloidal EPCs.

5.3 Electrode

ITO conductive glass is the most widely used electrode for the construction of a colloidal EPC system due to its high conductivity, high light transmittance, and ease of surface modification. Its conductivity can be optimized by controlling the thickness of the ITO layer and the doping ratio of tin during the magnetron sputtering. When a higher stability against acid/base is required, F-doped tin oxide (FTO) glass with lower transparency and conductivity can be an alternative electrode. The use of ITO or FTO as the top electrode of the EPC cell is beyond dispute. However, whether conventional electrodes, such as metal or carbon electrodes, can be used for the bottom electrode remains unsettled.

Unlike the adjustment of colloidal particles and dispersant, the surface modification of the electrode can effectively enhance the performance without altering the EPC's formulation. It makes electrode modification a stand-alone strategy, or an optional one that can be combined with formulation tuning, to enhance the EPC's performance. In this section, the structural and chemical modification of ITO electrodes will be discussed to clarify their contribution to the enhancement of stability, reversibility, and power consumption.

5.3.1 *Structural modification*

Needle-point discharge phenomenon indicates that it is easy to form a strong electric field at the sharp tip of a conductor when a voltage is applied to the tip and the counter electrode. Even though the size of the needle-like conductor is decreased to the nanoscale, it is still true to generate the enhanced electric field, which significantly improves the electrical responsibility of the EPC under the same voltage.

Yu et al. recently developed a structurally modified ITO electrode [72] for the EPC system and confirmed the above statements. Au, Cu, or Cu_2O nanoparticles were deposited onto the ITO electrode through ion-sputtering, electrodeposition, and colloidal coating. All the particle depositions lead to the formation of nanotip structures on the ITO electrode, which effectively enhances the charge injection and local electric field strength, and then improves the electrical response. As shown in Fig. 6, the SiO_2/(PCb-EG) EPC encapsulated in a regular ITO_+/ITO_- cell shows an onset voltage of 1.5 V and a voltage of 2.3 V to achieve a $\Delta\lambda$ of 120 nm. Its normalized color intensity declines to 0.4 as the voltage increases to achieve a lattice compression ratio above 10%. However, the same EPC encapsulated in an Au-ITO_+/ITO_- cell shows an onset voltage of 0.8 V, a voltage of 1.3 V for 120-nm blue shift, and a color intensity of 0.57 accordingly. Therefore, the Au modification to the ITO anode significantly lowers 47%/43% of the onset/working voltage, and retains a better color intensity at high voltages. Control experiments indicate that the above structural modification is most effective when the anode (attracting the particles) is deposited with an optimized amount of Au nanoparticles. Benefiting from the significant reduction in working voltage, particle deposition and electrochemical reaction are effectively suppressed on the electrode surface, which improves the long-term stability and reversibility of the response.

Later, Yu et al. found that the etching of ITO electrode with hydrofluoric acid (HF) [85] produces surface pits and circular edges due to the removal of ITO film, where the sharp edges also enhance charge injection and the local field strength to generate more sensitive electrochromic responses. Although the enhancement by HF etching is less effective than that from the nanotip structure,

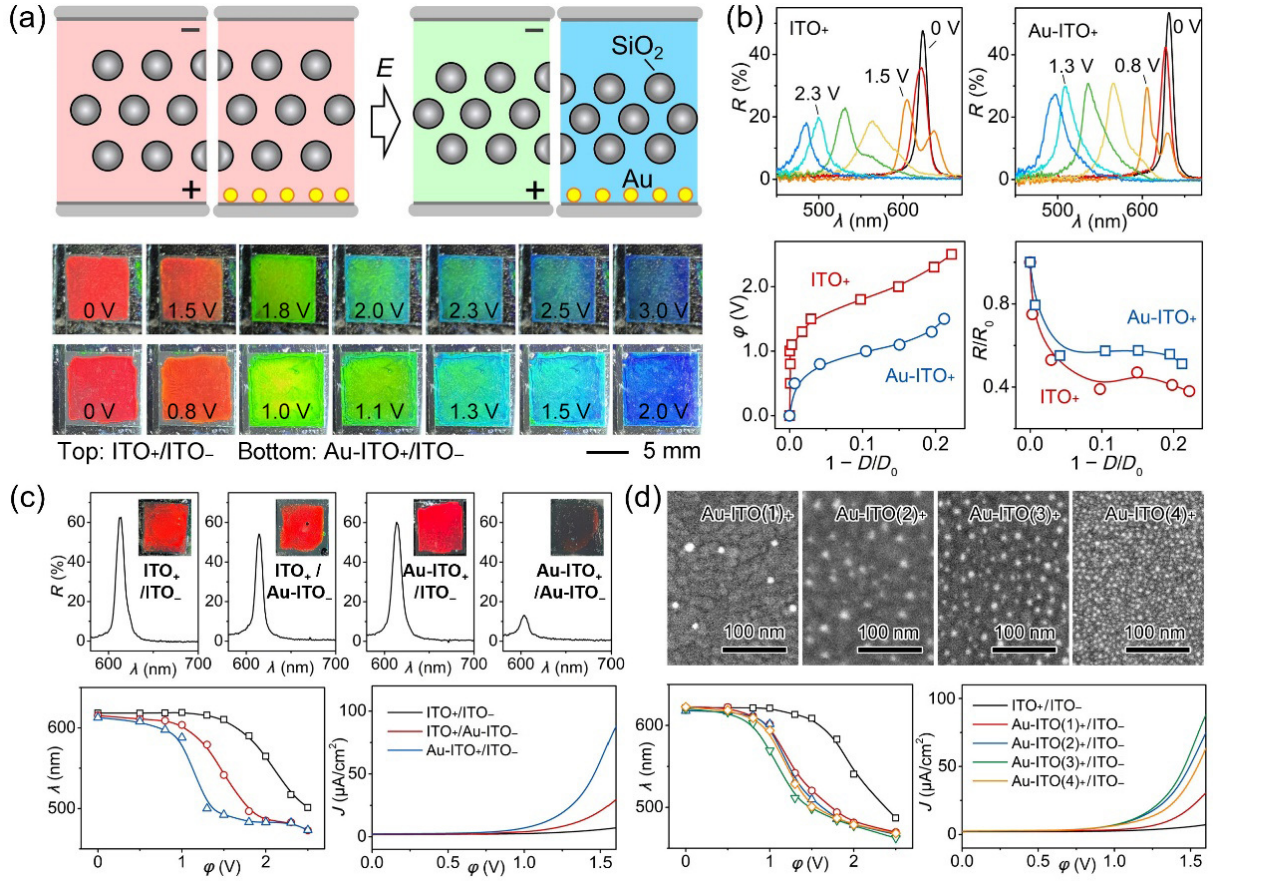

Figure 6 Performance enhancement by structural modification of the electrode. ((a) and (b)) Comparison of the electrochromic behavior, reflection spectra under different voltage, and the evolution of working voltage and color intensity with lattice compression ratio for the same SiO_2/(PCb-EG) EPC in a cell with ITO_+/ITO_- and Au-ITO_+/ITO_- as anode/cathode. (c) Reflection spectra and photos for SiO_2/(PCb-EG) EPC in the ITO_+/ITO_-, ITO_+/Au-ITO_-, Au-ITO_+/ITO_-, and Au-ITO_+/Au-ITO_- cells, and reflection wavelength and current density changed with increasing voltages. (d) SEM images of Au-ITO anodes with different amounts of Au deposition, and reflection wavelength and current density changed with increasing voltages. Reproduced with permission from Ref. [72], © Wiley-VCH GmbH 2023.

and excessive etching may disable the electrical response due to the removal of the whole conductive layer, it remains an alternative structural modification method considering the convenience of chemical etching and mechanical processing.

The structural modification of the electrode is especially favorable to produce a regioselective electrochromic response. For example, the degree of Au deposition or chemical etching in different regions of the ITO can be precisely controlled to achieve multi-level differentiated electrical responses. An electrically triggered anticounterfeiting tag based on this mechanism can gradually reveal the hidden pattern under increasing voltages.

5.3.2 *Chemical modification*

Unlike the structural modification, a chemical modification utilizes the deposition of a thin layer of active components, such as ion exchange resins, conductive materials, or charge-controlling polyelectrolytes, to inhibit the unwanted electrochemical reaction during electrical tuning or facilitate the colloidal assembly to achieve an enhanced color brightness.

Han et al. [33] reported an enhanced tuning stability for PS/H_2O EPC through the modification of the ITO electrode with ion exchange resins. In an aqueous EPC system, electrolysis of water is inevitable during the electrical color-tuning under a voltage of 1 to 4 V. The H^+ and OH^- ions generated at the electrode surface will gradually diffuse into the LPC lattice, which weakens the electrostatic interactions and harms the long-term stability of the optical response. As shown in Figs. 7(a)–7(c), a Nafion proton-exchange membrane (PEM) layer was coated onto the cathode to block the diffusion of OH^- ions generated at the cathode, whereas a Fumasep (FAS) anion-exchange membrane (AEM) layer was coated onto the anode to block the diffusion of H^+ ions generated at the anode. Such modification significantly increases stable switchings between red and green (100 nm) over 800 times, which is seldom realized by other EPC systems.

To address the influence of water electrolysis on the response stability of PS/H_2O EPC, Shim et al. [79] further studied the application of electrodes with a wider electrochemical potential window, including diamond-like carbon (DLC) and boron-doped diamond (BDD) electrodes. They prepared the DLC electrode by depositing 3-nm-thick DLC, 6-nm-thick Ti, and 3-nm-thick DLC on a Si wafer and used it as a cathode to fabricate the EPC cell. According to the cyclic voltammetry (CV) curve, the water electrolysis in the ITO-ITO cell begins at 1.5 V, while that in the ITO-DLC cell occurs over 2.6 V. When a maximum driving voltage of 3.2 V is applied to the EPC, the amount of generated H^+ and OH^- is significantly decreased for the latter case. Therefore, by introducing larger electrochemical potential window electrodes, the switching stability was appreciably enhanced through reducing the number of ions generated. The PS/H_2O EPC in the ITO-DLC and ITO-BDD cell shows a stable response for about 20 and 60 cycles

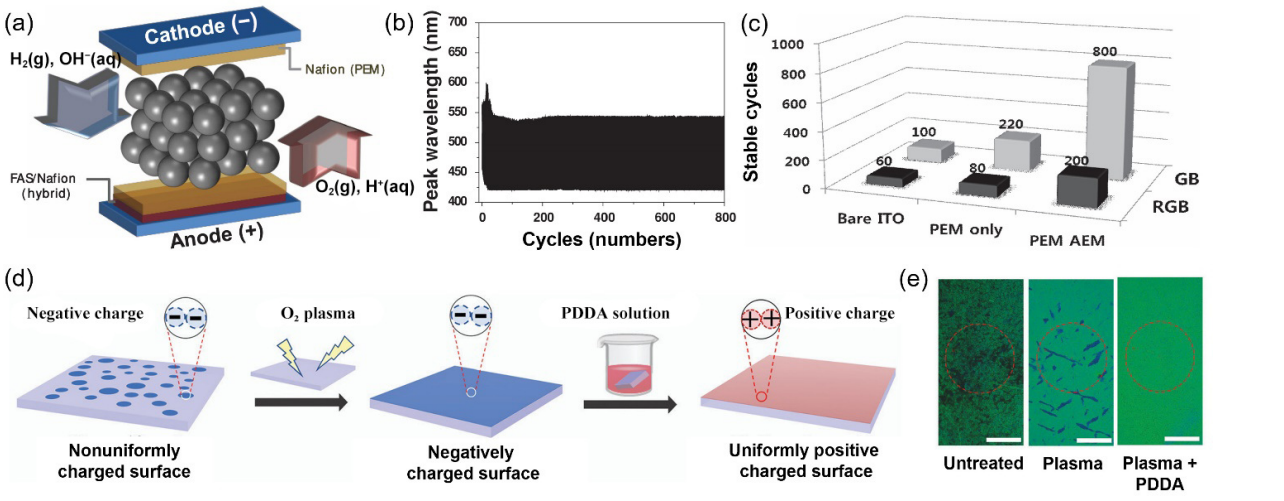

Figure 7 Performance enhancement by chemical modification of the electrode. (a) Blocking the diffusion of OH^- and H^+ ions in the PS/H_2O EPC system by coating the cathode and anode with ion-exchange resin membranes. (b) Highly reversible switching of reflection wavelength in 800 cycles due to electrode modification. (c) The maximum switching cycles of PS/H_2O EPC using ITO/ITO, (PEM-ITO)/ITO, and (PEM-ITO)/(AEM-ITO) as cathode/anode. (d) Preparation of a uniformly charged electrode by O_2 plasma treatment and PDDA modification. (e) Uniform color change under electric field due to electrode modification. ((a)–(c)) Reproduced with permission from Ref. [33], © WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim 2014. ((d) and (e)) Reproduced with permission from Ref. [67], © The Royal Society of Chemistry 2022.

when a high voltage is applied to achieve a wavelength shift of 150 nm. Furthermore, the replacement of the ITO cathode with a DLC or BDD cathode also avoids the reduction of In_2O_3 to metallic In, which is another factor for unstable response.

Song et al. [67] developed a Fe_3O_4@C/acetone EPC with bright uniform color, broad tuning range, and low driving voltage through plasma treatment of the electrode followed by modification with polydimethyl diallyl ammonium chloride (PDDA). As shown in Figs. 7(d) and 7(e), oxygen plasma treatment cleans the electrode surface to form a uniform negative surface charge, while the PDDA adsorption retains the charge uniformity and converts the surface to a uniform and positively charged one. Such chemical modification enhances the inductive effect of the glass substrate in colloidal assembly, as the coulombic interaction between the positively charged electrode and the negatively charged particles facilitates the growth of a short-range ordered colloidal lattice close to the electrode, which promotes the assembly in the outer layer and results in an increase of 110% in reflection intensity. Additionally, the charge distribution on the electrode is unified by the modification, which effectively reduces the formation of disordered regions, that is, the dark spots, and improves the color uniformity of the ERPC.

5.4 Driving waveform

As a functional device powered by electricity, the EPC cell eventually should be driven by a driving waveform of programmed voltages, which is similar to the commercial E-ink devices. During the electric tuning, the driving waveform dynamically adjusts the surface charge on the electrode and the electric field strength applied to the EPC, which further controls the electrophoretic behavior of colloidal particles and the electrochemical reactions on the electrode surface.

For most previous research, the EPC system is usually driven by a square waveform composed of a constant voltage and 0 V, where the voltage not only thermodynamically controls the structural color due to the balanced electrostatic interactions but also kinetically controls the electrophoretic speed to achieve that color change. Such highly coupled voltage control makes the EPC either slow response under low voltage or poor reversibility and stability under high voltage. However, through a carefully programmed driving waveform [44, 71], it is possible to address these issues, especially for those low-voltage EPCs.

In the research of Han et al. [33], they proved that the voltage driving methodology is an effective way to improve the tuning stability. As shown in Fig. 8, a square waveform switched between 3.2 and 0 V leads to a fast reflection wavelength change ($\Delta\lambda$) of 100 nm at the beginning. However, the $\Delta\lambda$ decreases to only 1/3 of the initial value after 250 cycles of switching. When a triangle waveform is applied to achieve the same color switching, the reflection slightly blueshifts, and the $\Delta\lambda$ remains the initial value, which shows much higher stability in electric tuning. The triangle waveform helps to shorten the time for EPC exposed to high voltage, which avoids the electrolysis of water to some extent.

Later, Yu et al. [72] also demonstrated that the driving waveform helps to accelerate the response speed. During a lattice compression process, an "over potential" modulation can effectively accelerate the electrical response speed in EPC tuning as the electrophoretic mobility of colloidal particles is directly proportional to the applied voltage. A "step rising" modulation is also an effective method for fast response during lattice compression. The continuously strengthened Coulombic force avoids the destruction of the ordered lattice at the beginning and provides adequate strength to compress the lattice later. For a SiO_2/(PCb-EG) EPC changing from red to green under 1.3 V, the application of a short period of "over potential" at 1.5 V decreases the response time from 42 to 30 s, and a step rising voltage further decreases the response time to 5 s.

In a lattice expansion process, a "reverse potential" modulation can shorten the color recovery time, as the reversed Coulombic force facilitates the lattice expansion from the highly viscous compressed state. Meanwhile, a "pulsive potential" modulation is found to be effective to avoid the compression of the already expanded lattice towards the opposite electrode, as the pulse numbers can be precisely controlled. For the same SiO_2/(PCb-EG) EPC [72] recovering from green to red, the application of a period

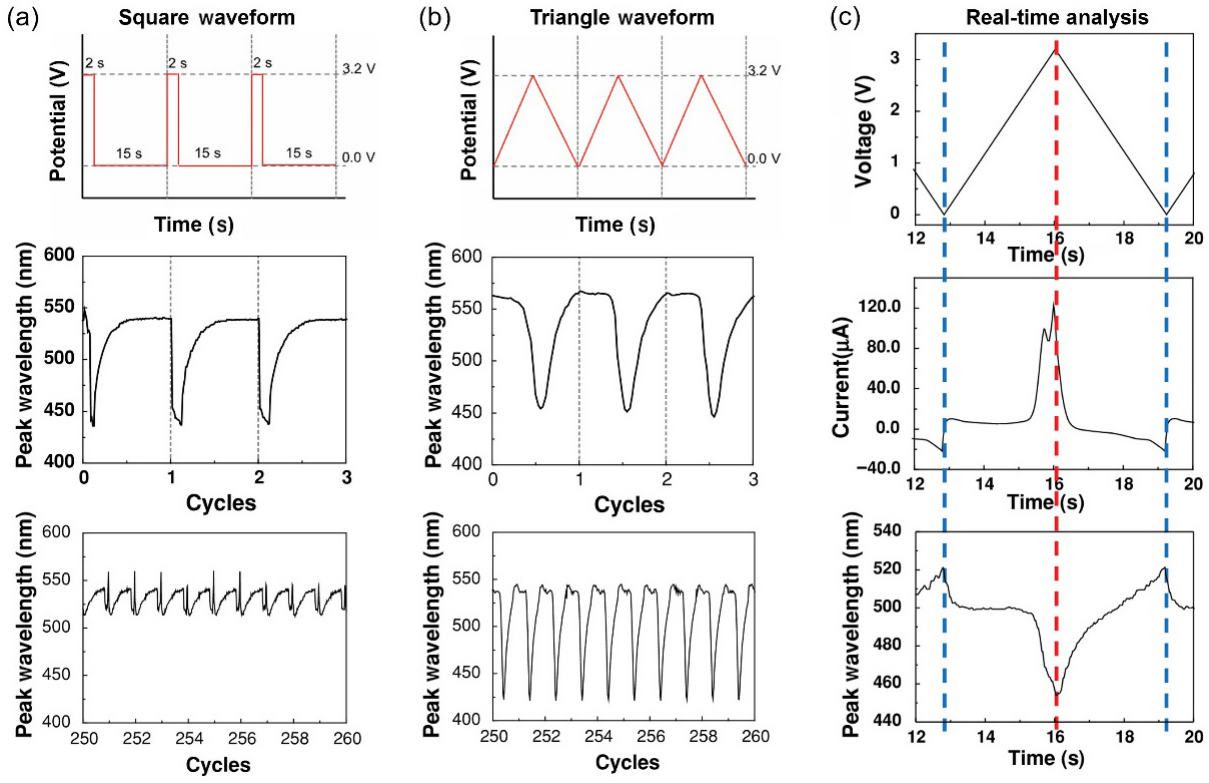

Figure 8 Performance enhancement by programming of driving waveform. Reflection wavelength change of PS/H_2O EPC under the tuning: (a) a square waveform and (b) a triangle waveform in 260 cycles. (c) Real-time analysis of the current flow and the reflection wavelength change with the applied voltage. Reproduced with permission from Ref. [33], © WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim 2014.

of "reverse potential" at −1.5 V decreases the response time from 44 to 20 s. When the reverse potential is combined with pulsive modulation, the response time further decreases to 10 s.

6 Outlook and perspectives

This review article summarizes the photonic crystal materials, the working mechanism, the evaluation standards, and the strategies of performance enhancement for the colloidal EPC system. Since the first report of colloidal EPC in 2010, significant progress has been achieved in the following years. More liquid PC materials composed of different colloidal particles and dispersants are synthesized to construct EPC systems with improved or unique responses. The electrochromic mechanisms, such as "lattice compression and expansion", "colloidal concentration and redispersion", and "lattice collapse and recovery", are well studied and verified with solid experimental results. The electrochromic performances in the aspect of color tuning range, working voltage, response sensitivity, power consumption, response speed, color intensity, reversibility, and stability are improved to varying degrees by different methods.

Although great improvement has been achieved for colloidal EPCs, there are still many challenges for their development and commercial application. First of all, large-scale manufacturing of colloidal EPC materials with low cost and high quality is the basis for all research and industrialization. Currently, SiO_2 particle is still the best choice for the construction of colloidal EPC, considering its large-scale synthesis, narrow size distribution, precisely tunable size, highly charged surface, and good stability in organic solvents. A typical SiO_2/PCb EPC with f_{SiO_2} of 20% can be prepared in a laboratory on a liter scale with a reagent cost of 300 CNY/L. Fabrication of colloidal EPC based on other particles is a direction for future development and performance enhancement, but fundamental research in synthesis is required to reduce the cost and improve the quality of colloidal particles first.

Integration with existing optoelectronic platforms is also a challenge to accelerate the development of colloidal EPC. It is known that both the electrophoretic display (EPD) and the colloidal EPC work through particle electrophoresis under electric field. This enables the colloidal EPC to be fully compatible with the previous optoelectronic devices and techniques developed for EPD. Nevertheless, inherent differences between them require extensive work to achieve the integration. A micro cup electrophoretic cell instead of a polymer microcapsule might be a suitable way to encapsulate colloidal EPC materials, because the ordered structure of EPC is easily affected by the polymerization process. Integrated circuit (IC) chips and driving waveforms need to be developed to precisely control the electrical response of the EPC, as the electrophoresis should be accomplished without disturbing the ordered structure in most cases.

Furthermore, the EPC performance needs to be improved significantly to meet the requirements of commercial products. For instance, a rapid electrochromic response at the millisecond level is a big concern, considering the working voltage cannot be largely enhanced to accelerate the colloidal electrophoresis. Highly reversible and stable exhibition of structural color is also difficult to achieve, as the unwanted electrochemical process or physical changes may occur during electric tuning. Pathway-independent color tuning has not been realized yet, as the same voltage sometimes leads to different colors in different tuning pathways. Furthermore, an optimization may influence two performances oppositely, so that a more precise and balanced control is required.

Nevertheless, the colloidal EPC apparently has advantages in low consumption compared to the liquid crystal-based devices (Table 2). It provided a full-color display instead of black/white or transparent/opaque switching compared to the inorganic electrochromic oxides and conventional monochromic EPD. As a strong competitor to color EPD based on four-colored particles, the colloidal EPC has advantages in manufacturing cost, color control, and response time because the preparation and electric tuning of a single material is easy to realize. To fully leverage its advantages, more investigations are required in EPC's material design, electrophoretic cell fabrication, device packaging, and waveform programming, We believe that significant breakthroughs will be achieved by the combined efforts from researchers in different fields, which lays down a firm basis for its potential application in electrophoretic color display, smart window, adaptive camouflage, and optical devices.

Data availability

Not applicable

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Declaration of competing interest

All the contributing authors report no conflict of interests in this work.

Author contribution statement

S. Y. Y.: Literature investigation, writing – original draft. W. H. Z.: Literature investigation. X. M. L.: Literature investigation. X. D. L.: Literature investigation. J. P. G.: Project administration, funding acquisition, writing – review & editing. All the authors have approved the final manuscript.

Use of AI statement

None.

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Table 2 Summary of the electrochromic mechanisms, performance, advantages, and limitations for colloidal EPCs, inorganic electrochromic oxides, liquid crystal displays, and EPDs

Type (typical material)	Electrochromic mechanism	Color tuning mode	Driving voltage (V)	Response time (s)	Advantages	Limitations
Colloidal EPCs (SiO_2/PCb)	Colloidal electrophoresis-induced lattice change in photonic crystal	Full-color tuning or brightness tuning	0–4	0.5–5	1. Broad color tunability 2. High color saturation 3. Antiglare structural color 4. Low consumption	1. Slow response 2. Unsatisfactory stability and reversibility (currently)
Inorganic electrochromic oxides (WO_3)	Ion intercalation/ deintercalation-induced variation in transmittance and reflectance	Transparent/opaque switching	0–5	0.1–10	1. Good stability and long lifetime 2. Easy fabrication 3. Good compatibility	1. Low color purity and brightness 2. Single-color switching
Liquid crystal displays (4-cyano-4'-pentylbiphenyl)	Polarization and transmittance controlled by the orientation of liquid crystal	Black/white switching (color tuning, combined with filter)	1.5–6	0.001–0.02	1. Mature technology for wide application scope 2. High resolution 3. Fast response 4. Compatible to flexible devices	1. Relies on back light and color filters 2. Relatively high consumption 3. Light leakage
Monochromic EPDs (TiO_2-carbon black)	Selective electrophoretic movement of charged particles in a dielectric fluid towards a specific electrode	Black/white switching (color tuning, combined with filter)	−15–15	0.1–3	1. Low consumption 2. Eye-friendly 3. Wide viewing angle	1. Low refresh rate 2. Screen flickering during full refresh 3. Unsaturated color when using color filter
Color EPDs (polymer particles encapsulating organic dyes)	Precise vertically positioning of four-colored particles with different charges through electrophoresis	Full-color tuning	−30–30	10–30	1. Low consumption 2. Eye-friendly 3. Wide viewing angle 4. Saturated color	1. Ultralong switching time 2. Complicated driving waveform

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