REVIEW

Circularly polarized luminescent self-organized helical superstructures: From materials and stimulus-responsiveness to applications

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Abstract
Circularly polarized luminescence (CPL) has gained considerable attention in various systems and has rapidly developed into an emerging research field. To meet the needs of actual applications in diverse fields, a high luminescence dissymmetry factor (g_lum) and tunable optical performance of CPL would be the most urgent pursuit for researchers. Accordingly, many emerging CPL materials and various strategies have been developed to address these critical issues. Emissive cholesteric liquid crystals (CLCs), that is, luminescent self-organized helical superstructures, are considered to be ideal candidates for constructing CPL-active materials, as they not only exhibit high g_lum values, but also enable flexible optical control of CPL. This review mainly summarizes the characteristics of CPL based on CLCs as the bulk phase doped with different emitters, including aggregated induced emission molecules, conventional organic small molecules, polymer emitters, metal–organic complex emitters, and luminescent nanoparticles. In addition, the recent significant progress in stimulus-responsive CPL based on emissive CLCs in terms of several types of stimuli, including light, electricity, temperature, mechanical force, and multiple stimuli is presented. Finally, a short perspective on the opportunities and challenges associated with CPL-active materials based on the CLC field is provided. This review is anticipated to offer new insights and guidelines for developing CLC-based CPL-active materials for broader applications.

KEYWORDS
amplifying dissymmetry factor, cholesteric liquid crystal, circularly polarized luminescence, self-organized helical superstructure, stimulus-responsiveness

INTRODUCTION

Chirality is a basic attribute of matter and is also widespread in nature at various hierarchical levels, from the subatomic and molecular scales to the supramolecular, nanoscopic, macroscopic, and galactic scales. Based on the interaction between chirality and electromagnetic waves, various optical effects have emerged, especially the well-known circular dichroism (CD) and circularly polarized luminescence (CPL). CPL, which contains the differential emission intensity of left- and right-handed CP light, can reflect the structural information in the excited state of chiral luminescent materials or systems. In recent years, functional materials with CPL characteristics have attracted considerable interest owing to their fascinating application prospects in three-dimensional (3D) optical imaging, chiral polymerization, chemical sensors, tunable lasers, photoelectric devices, and so on. To evaluate the performance of CPL-active
SCHEME 1 Generation and regulation of circularly polarized luminescence (CPL) induced by the self-organized helical superstructure

with materials, the dissymmetry factor of luminescence ($g_{\text{lum}}$) is a crucial index, which is given by $g_{\text{lum}} = \Delta I/I = 2(I_L - I_R)/(I_L + I_R)$, where $I_L$ and $I_R$ are the intensities of the pure left- and right-handed CPL, respectively. The value of $g_{\text{lum}}$ ranges from +2 for ideal left CPL to −2 for ideal right CPL.\[15\] To date, many emerging CPL-active materials, including lanthanide metal complexes, organic molecules, natural macromolecules, synthetic polymers, self-assembly systems, and chiral liquid crystals (LCs), have been reported.\[16–35\] In terms of practical application, the development of an appropriate CPL material with a high degree of polarization has become urgent.\[36,37\] Nevertheless, it is difficult for most CPL materials to provide high asymmetry factors to meet the needs of some optoelectronic applications. Recently, more attention has been paid to the many merits of cholesteric LCs (CLCs), that is, self-organized helical superstructures. As expected, CLCs would provide new opportunities for developing CPL-active materials with remarkable properties because the self-organized helical superstructures of CLCs possess many unique optical properties, including optical rotation, CD, and selective reflection.

Functional LC materials with chiral architectures have been widely investigated for use in diverse applications, where the chirality endows these unique soft materials with unprecedented optical, photonic, and mechanical properties.\[38–50\] The chiral LCs contain diverse types, typical examples of which include CLCs, blue phases, and chiral smectic LCs. Among them, CLCs are known to have chirality at the molecular level and non-superimposable helical superstructures at the macroscale level. In the CLC phase, the molecules exhibit spatial helicoidal orientations along the helical axis, as shown in Scheme 1. In general, the helicoidal molecular arrangement is characterized by two important parameters: twist handedness and helical pitch ($P$).\[51–54\] Specifically, $P$ is employed to express the distance at which the director of the CLC rotates a full 360°. According to Bragg’s law, CLCs exhibit selective reflection of CP light. At normal incidence, the maximum wavelength of the reflected light is defined by $\lambda = nP$, where $n$ is the average refractive index of the LCs. In CLC-constructed CP luminescent materials, the CPL performance is also closely related to the above optical properties of CLCs. There are two possible mechanisms through which $g_{\text{lum}}$ is amplified in CLCs. In the first mechanism, the optical rotation from the helical superstructure of the CLCs gives rise to a high CPL polarization degree when the emission peak does not overlap with the reflection band. Thus, the handedness of the CLCs mainly determines the polarized direction of the obtained CPL. The second mechanism is based on CD: CPL with the same sense as the helical twist of the helical superstructure is totally reflected when the emission peak coincides with the reflection band, whereas CPL with the opposite handedness is transmitted. Consequently, an ultrahigh $g_{\text{lum}}$ could be obtained in this case.\[55\] In addition, it is worth mentioning that cellulose nanocrystals (CNCs)-based self-organized helical superstructures as representative lyotropic CLCs have attracted considerable attention for use as the matrices of CPL-active materials in recent years. This review focuses on recent advances in CLCs with self-organized helical superstructure for constructing CPL-active materials.

Regarding the generation of CPL, both chirality and luminescence are necessary factors. At present, there are two typical methods for designing and constructing CPL materials. One approach is to introduce a chirality moiety into fluorophores to obtain CPL-active materials. Another universal strategy is doping fluorescent dyes into chiral systems.\[5,16\] In general, introducing chiral or achiral luminescent materials into achiral LCs or chiral LCs, respectively, is a typical method of fabricating CLC-based CPL materials. To date, many achiral or chiral luminescent materials have been widely used as emitters to construct CPL-active materials based on CLCs, such as organic fluorescence...
molecules, luminogens with aggregation-induced emission (AIE) property, carbon dots (CDots), upconversion nanoparticles (UCNPs), quantum rods (QRs), conjugated polymers, semiconductor quantum NCs, and inorganic perovskite NCs (PKNCs). The use of different types of emitters has greatly enriched the CPL-active materials based on CLCs, endowing the constructed system with diverse properties. For example, CLC-based CPL materials constructed from AIE molecules simultaneously exhibit high photoluminescence quantum yields and large dissymmetry factors compared to those of the conventional fluorescence dye in CLCs.[21] The following sections will discuss CLC-based CPL-active materials based on different types of fluorescence dopants. One means of fabricating cholesteric conjugated polymers or microspheres with CPL characteristics is to add a chiral dopant into an achiral π-conjugated polymer or to introduce a chiral moiety into the polymer side chains at the molecular level, and another is to use CLCs as asymmetric polymerization solvents to fabricate helical polymers. These different preparation strategies provide infinite possibilities for the construction of CLC-based CPL-active materials.

Stimulus-responsive CPL materials with CPL properties that can be regulated by altering external stimuli have received tremendous attention due to their potential use in some specific applications, especially in security encryption and sensor devices.[56] Recently, stimulus-responsive CPL-active materials based on CLCs have been favored by researchers, and many remarkable achievements have been reported. There are diverse methods of dynamic CPL switching in CLC media, which can be attributed to the stimulus-responsive nature of the CLCs. Up to present, many external stimuli have been used to switch CPL properties in CLC media, including light, temperature, pH, solvent, electric field, and force. In these cases, the intensity, wavelength (i.e., color if it is at visible region) or handedness inversion of CPL signals can be switched in the constructed CLC system. From the perspective of the working mechanism, CPL switching in an emissive CLC system has been realized by either tuning molecule structures to change the related helical assembly behaviors or tuning external interactions between chiral and achiral components/moieties. Based on their excellent switchable properties, CLC-based CPL-active materials possess considerable potential for applications in 3D displays, anti-counterfeiting, encryption, and smart sensors.

Duan et al. recently published an excellent review summarizing the properties of CPL-active CLCs based on small molecules, CPL-active polymer CLCs, and CPL in CNC-based helical superstructures.[55] However, the current review is the first to focus on the introduction of various emitter-based CLCs with tunable CPL characteristics, with the aim of providing a more comprehensive summary of the entire research area. Specifically, it highlights recent progress in CLC-based CPL-active materials doped with different types of emitters, such as AIE molecules, conventional organic small molecule emitters and polymer emitters, metal–organic complex emitters, and luminescent nanoparticles. It also describes methods of dynamically switching CPL in emissive CLCs, including approaches using light, temperature, humidity, and electric fields. In addition, it discusses the fascinating properties of CLC-based CPL systems used to construct smart photonic materials for 3D display, sensing, and encryption applications. In particular, some very recent important studies that have influenced the development of this captivating field are highlighted. The review concludes with a brief perspective on some major challenges and opportunities for the future development of CLC-based CPL materials.

### CPL GENERATION

CPL can be generated by the excited states of chiral molecules or assemblies. To meet practical application requirements, both strong signals and large $g_{\text{lum}}$ values are indispensable for CPL-active materials. Extensive efforts have been devoted to the development of high-performance CPL materials, which have generally been focused on enhancing the luminescent ability or improving the capacity of chiral separation. In the latter respect, CLCs are considered as excellent hosts for amplifying the chirality and can exhibit quite high $g_{\text{lum}}$ values because of the highly ordered helical arrangement of the LC molecules.[57–59] This section will focus on CLC-based CPL systems constructed with different luminogens.

#### Organic fluorescent emitters

Small molecular organic dyes seem to be suitable candidates for CPL emitters because their properties can be tailored through reasonable structural design, such as tunable emission wavelength and high fluorescence quantum yield.[60] Developing high efficiency fluorescent dyes is essential to obtain strong CPL emission. Tang et al. discovered a new type of molecules that showed intense fluorescence in the aggregated state, and this phenomenon was termed the AIE effect.[61,62] These distinctive AIEgens are expected to underpin the development in the CPL field with enormous potential. In the constructed CPL-active materials based on CLCs and other chiral LCs, CPL can be generated by three types of emitters: AIE-active achiral molecules, AIE-active chiral molecules, and other traditional non-AIE fluorescent dyes.

##### AIE-active achiral molecules

Emissive CLCs constitute an excellent medium to engender CPL. In general, CLCs can be obtained in two ways: from the self-assembled behavior of cholesterol-based derivatives and from the introduction of chiral dopants into nematic LCs, which can induce LC molecules to form helical superstructures. The simultaneous doping of fluorescent molecules is a relatively easy method of obtaining emissive CLCs. Cheng et al. first reported super-high $g_{\text{lum}}$ values induced by supramolecular self-assembly in LCs (Figure 1A).[63] They developed ternary LC mixtures by incorporating a pair of chiral dopant enantiomers (guest 1) and four different achiral AIE-active dyes (guest 2) into host E7, called AIE-CLCs. The four types of AIE-CLCs exhibited strong CPL with $g_{\text{lum}}$ values ranging from 0.97 to 1.42. It is worth noting that all of the AIE-CLCs showed higher $g_{\text{lum}}$ values than CLCs (without AIE-dyes), indicating highly ordered helical π–π stacking between the binaphthyl moieties of guest 1 and the phenyl group of guest 2, as well as supramolecular self-assembly between guests 1 and 2 in host E7.
Figure 1 (A) Strong circularly polarized luminescence (CPL) with super-high $g_{\text{lum}}$ values induced by supramolecular self-assembly consisting of chiral dopants, achiral aggregation-induced emission (AIE)-active dyes and nematic liquid crystals (LCs). Reproduced with permission. Copyright 2019, Royal Society Chemistry. Ref. [63] (B) Ultrasstrong red CPL promoted by intermolecular energy transfer and chirality induction of the emissive ternary chiral LCs (T-CLCs). Reproduced with permission. Copyright 2021, American Chemical Society. Ref. [64] (C) Structurally colored polymer films with CPL emission prepared by the photopolymerization of cholesteric liquid crystal (CLC) mixtures. Reproduced with permission. Copyright 2021, Royal Society Chemistry. Ref. [65]

Recently, Cheng et al. reported a kind of CPL material with a high $g_{\text{lum}}$ of up to 1.51 through intermolecular energy transfer and chirality induction from the supramolecular self-assembly of CLCs with chiral AIE molecules (guest A) serving as both chiral inducers and energy donors, and the achiral AIE acceptors (guest B) as CPL emitters were doped into the nematic LC E7 host to form an emissive ternary chiral LC (T-CLC) (Figure 1B). [64] Guests A and B showed excellent compatibility with host E7 because they all contained cyano and phenyl groups. Interestingly, the acceptor could gather energy from the chiral donor and emit a red CPL signal; therefore, an ultrastrong CPL signal was obtained by further amplification through the Förster resonance energy transfer process in the T-CLCs.

In addition to small molecular LC systems, LC polymers with CPL properties have attracted much attention owing to their unique stabilization and various functionalities. Yang et al. reported a CLC-based structurally colored polymer film accompanied by CPL emission, using a facile method of photopolymerization of CLC mixtures (Figure 1C). [65] The photopolymerizable CLC mixtures were composed of C6M and RM105 as the reactive LCs, BDK as the photoinitiator, R/S5011 as the chiral dopant, and a star AIEgen tetraphenylethylene (TPE), which was selected as the CPL emitter. The polymer films were synthesized by photocrosslinking reactions at the CLC phase and displayed reflection colors from red to purple according to the weight ratio of the chiral dopants. Notably, all these polymer films showed CPL signals at approximately...
485 nm with absolute $g_{\text{lum}}$ values in the range of 0.53–0.58, indicating that the helical pitches of the supramolecular assemblies did not have much influence on the CPL intensity.

**AIE-active chiral molecules**

Since Tang et al. first synthesized a chiral AIE molecule containing luminogenic silole and chiral sugar moieties, which showed good CPL performance in the solid state, various AIE chiral fluorescent molecules have been reported as CPL emitters. For instance, Cheng et al. reported two pairs of AIE-active chiral fluorescent dyes (R-/S-1, R-/S-2) with similar molecular structures, of which R-/S-2 had a higher quantum yield in the 5CB host because of the strong intramolecular charge transfer effect (Figure 2A) [67]. CPL-emitting CLCs were obtained by doping the chiral AIEgens into the 5CB host, designated as CLCs-1 and CLCs-2 respectively, and
they showed different CPL signals. R-/S-2 exhibited a 3.7-fold $g_{\text{lum}}$ value as high as 0.37 compared to R-/S-1 in 5CB, which was attributed to the stronger D-A structure and more planar conformation of R-/S-2.

Chiral amplification in CLCs is a well-known strategy; however, the underlying mechanism remains ambiguous. Tang et al. explored the coassemblies of chiral AIE molecules and LCs at the nanoscale for the first time (Figure 2B).[68] The TPE derivatives kept CD silent in tetrahydrofuran solution, even in a suspension. By doping AIEgens into 5CB, considerably enhanced CD and CPL signals were detected. To investigate the mechanism of chiral amplification, atomic force microscopy was used to reveal the formation of the coassemblies of AIEgens and 5CB at the nanoscale. A pearl-necklace morphology of the coassemblies could be clearly observed, with chiral TPE derivatives internally forming the helical fibers and 5CBs assembled along the helical fibers of the AIEgens in a similar helical arrangement. The theoretical calculations also supported this observation. These results provide crucial experimental and theoretical evidence for chiral amplification in CLCs and will facilitate the design of supramolecular systems composed of chiral additives and nematic LCs.

Non-AIE fluorescent molecules

Duan et al. demonstrated signal transmission encryption based on CPL engendered by dye-doped CLCs (Figure 3A).[69] Incorporating R/S-811 as a chiral dopant and C6 as a CPL emitter into the commercially available LC medium SLC1717 induced the formation of helical superstructures of CLCs. When the emission of C6 was located at the center of the photonic bandgap (PBG), the CLCs exhibited the highest $g_{\text{lum}}$ ascribed to the PBG effect, which inhibited CPL with handedness consistent with CLCs. The influence of the LC cell thickness on the CPL was further explored. To avoid irregular LC orientations caused by the uneven thickness, the authors used a double-layer LC cell, where each layer had a thickness of 29 mm to replace the LC cell with the same overall thickness of 58 mm. An interesting finding was that the $g_{\text{lum}}$ value of the CLCs in the double-layer cell was dramatically amplified from 1.07 to 1.52. Such an outstanding CPL can be used in information transmission. By rotating the quarter-wave plate and linear polarizer, the output signals of the CLCs changed between sinusoidal and square forms, whereas the modes for polarization detection both showed a sine wave when the achiral LC contained only C6 as the sample. Therefore, the square wave signal generated by the input CPL of the CLC can be considered as an encrypted signal.

Recently, CP lasers have made encouraging progress in the field of 3D laser displays. Zhao et al. developed a complex but ingenious design to form helical network polymers using chiral LCs as asymmetric polymerization solvents (Figure 3C).[70–72] The CLC template was prepared by adding chiral dopants into a nematic LC host, and the LC mixture contained two kinds of methacrylate monomers, one with a liquid crystalline unit and the other with a fluorescent moiety, as well as a triacrylate-based cross-linker and a photoinitiator. The helical network polymer was synthesized through photo-cross-link polymerization upon irradiation with ultraviolet (UV) light and then soaked in chloroform for 72 h to separate the unpolymerized molecules and obtain a well-morphological polymer film. The helical network polymers showed fingerprint and spiral morphologies similar to those of the CLCs, which supported blue CPL emission.

In addition, earth-abundant and renewable CNCs that possess chiral glucose units enable self-assembly in suspensions and form inherent left-handed helical superstructures with chiroptical properties.[74–76] In a broad sense, the self-assembly of CNCs can be considered as left-handed CLCs, and they can serve as hosts for various nanomaterials owing to their excellent compatibility. Considerable right-handed CPL signals can be generated by CNC templates when coassembled with fluorescent dyes.

Xu et al. exploited the circular polarization potential of CNCs to enable right-handed CPL because of its PBG and left-handed helical sense (Figure 3D).[77] The right-handed CPL emission was realized by incorporating different achiral fluorescence dyes (i.e., M1 and M3) into CNC films. The CPL positions and $g_{\text{lum}}$ values of the CNC films changed with PBG. CNC-M1-1 and CNC-M1-2, which featured the maximum spectral overlap, exhibited the highest $g_{\text{lum}}$ values of $-0.68$ and $-0.55$ respectively, which could be attributed to the forbidden propagation of the left-handed CPL emission within the PBG. Furthermore, strong right-handed CPL values of CNC-M1-3 and CNC-M1-4 with corresponding $g_{\text{lum}}$ values of $-0.51$ and $-0.57$ were also observed because of the enhancement at the photonic band edge (PBGedge), although their emission bands were further away from the PBG. They also demonstrated a “butterfly” pattern consisting of both M1 with emission inside the PBG and M3 with emission outside the PBG. Such a design could regulate the right-handed CPL induced by M1 with the use of polarization, so an orange pattern was observed with the left-polarizing filter, whereas a beige butterfly was observed with the right-polarizing filter.

Owing to their remarkable emissive efficiency, diverse structural designs, and excellent compatibility with the host, organic fluorescent molecules have been widely applied in CPL-active materials. As discussed, CPL-emitting systems can be constructed via two main approaches: (1) doping achiral fluorescent molecules as CPL emitters into helical superstructures/assemblies, such as CLCs, LC polymers using CLCs as templates, and CNCs; (2) introducing chiral fluorescent molecules into nematic LCs, which induces long-range ordering of the cholesteric structures, where the chiral fluorescent molecules serve as both luminogens and stereocircum vision theory, such CP laser arrays with orthogonal circular polarizations can provide a 3D sense to the brain.

Polymerization of fluorescent monomers with polymerizable functional groups is an effective means of obtaining luminescent LC (LLC) polymers.[70–72] On the other hand, the helical structure of small-molecule CLCs can serve as a polymeric template for one-handed LC polymers. Akagi et al. developed a complex but ingenious design to form helical network polymers using chiral LCs as asymmetric polymerization solvents (Figure 3C).[73] The CLC templet was prepared by adding chiral dopants into a nematic LC host, and the LC mixture contained two kinds of methacrylate monomers, one with a liquid crystalline unit and the other with a fluorescent moiety, as well as a triacrylate-based cross-linker and a photoinitiator. The helical network polymer was synthesized through photo-cross-link polymerization upon irradiation with ultraviolet (UV) light and then soaked in chloroform for 72 h to separate the unpolymerized molecules and obtain a well-morphological polymer film. The helical network polymers showed fingerprint and spiral morphologies similar to those of the CLCs, which supported blue CPL emission.
chiral dopants. In particular, AIE-active molecules overcome the aggregation-caused quenching (ACQ) effect of traditional organic dyes, benefiting the development of CPL materials with enhanced intensity and high $g_{\text{lum}}$ in the aggregate state.

**Luminescent LC (LLC) molecules**

LLCs combine photophysical behavior with anisotropy properties and have received tremendous attention in display and imaging technology.\(^{78,79}\) $\pi$-conjugated polymers with rod- or disk-like cores and flexible alkyl chains can be regarded as typical small-molecular LLCs and exhibit various LC mesophases, such as columnar, smectic, and nematic phases.\(^{19}\) $\pi$-conjugated polymers are also promising LLC materials. Driven by the chiral components, LLCs can spontaneously form helical structures, which provide good media for CPL emission. The following sections will introduce CPL-active materials based on LLCs from the two aspects of small molecules and polymers.

**LLC based on small molecules**

A straightforward and powerful means of obtaining LLCs is to link chromophores with LC moieties. Tang et al.
constructed a series of CPL-active molecules with AIE properties by combining dicyanodistyrylbenzene (DCS) units with cholesterol (Figure 4). All DCS-cholesterol derivatives (DCS-Chols) featured large quantum yields of fluorescence up to 73.7% in a condensed state and were completely free of the ACQ effect. The helical molecular packing in the LC states endowed a strong CPL signal with a high $g_{\text{Lum}}$ of 0.11, which was 157-fold higher than $g_{\text{Lum}}$ in solution. Interestingly, the handedness of CPL was dependent on the length of the alkyl spacers between DCS and cholesterol. DCS-Chols with five methylene units (α-5C, β-5C) exhibited right-handed CPL, whereas others containing eight methylene linkers (α-8C and β-8C) emitted left-handed CPL.

**LLC polymers**

Helical-conjugated polymers can form three types of π-stacked structures, including intrachain-twisted, intrachainspiral, and interchain-helical structures. The intrachainspiral structure of LLC polymers is usually constructed by introducing chiral moieties into the side chains at the molecular level. Akagi et al. synthesized various types of helically π-stacked thiophene-based copolymers by introducing chiral alkoxy carbonyl substituents into their side chains, which showed liquid crystallinity at high temperatures (Figure 5A). These polymers exhibited RGB-colored CPL in both solution and films with $g_{\text{Lum}}$ values on the order of $10^{-3}$ and $10^{-2}$, respectively. The higher $g_{\text{Lum}}$ value of the polymer films was attributed to the strengthening of the helical π-stacking in the solid state. An interesting finding was that the films annealed at LC temperature for a while generated an enhanced CPL with a $g_{\text{Lum}}$ value of up to $10^{-1}$, because the self-ordering hierarchical arrangement in CLCs promoted large interchain interactions. In addition, a white CPL across most of the visible region was obtained by mixing RGB trichromic polymers in an appropriate ratio.

Another interchain-helical structure of LLC polymers can generate amplified CPL, because it enables the polymers to self-assemble into an interchain helix. Akagi et al. reported one case in which chirality in disubstituted liquid crystalline polyacetylene derivatives was induced by means of either chiral side chains or the use of chiral dopants (Figure 5B). PA2 and PA3 possessing stiff polymer structures showed lyotropic nematic LC phases in organic solvents, whereas PA1 with the alkyl chain did not exhibit lyotropic LC behavior, which was likely attributed to the higher solubility. Furthermore, CPL could only be observed in the LC phase and remained CPL-silent both in solution and in cast-film LLCs. Taking PA2 as example, (R)-(S)-PA2 displayed a lyotropic CLC phase when dissolved in organic solvents, whereas (rac)-PA2 doped with chiral (R)-(S)-D1 also spontaneously aligned to form a helical structure in the CLC phase. Thus, the highly ordered lyotropic CLC generated intense CPL with a high $g_{\text{Lum}}$ value on the order of $10^{-1}$.

Through intermolecular interactions, the interchain-helical structure spontaneously formed by chiral LLCs can be further assembled into other nanosized or micrometer-scale shapes. Akagi et al. also reported an interchain helically π-stacked assembly in which cationic LLC polymers (PPP) were combined with anionic chiral dopants (BNP) by electrostatic and π-π interactions, which self-organized into a spherulite with blue CPL (Figure 5C). Each unit structure of the polymer assembly consisted of two PPP repeating units and one BNP molecule according to a stoichiometry estimation experiment. The polymer spheres in solution showed a Maltese cross pattern, indicating that the polymer spherulite was composed of crystalline lamellae (ionic-conjugated polymers) and interspaced noncrystalline sites. It could be regarded that the polymer assemblies further aggregated to form
spherulites bearing the interchain helical $\pi$-stacking structure, and the spherulites exhibited CP blue luminescence with $\delta_{\text{lum}}$ of $-2.4 \times 10^{-2}$ for PPP-(R)-BNP and $+3.1 \times 10^{-2}$ for PPP-(S)-BNP.

Very recently, Yamamoto et al. reported anisotropic CPL generated by solid microspheres with a twisted bipolar interior for the first time (Figure 5D).\cite{86} These solid microspheres were constructed using $\pi$-conjugated LLC polymers with chiral side chains and formed during the slow precipitation of the isotropic solution. As the solvent evaporated, the liquid droplets passed through the cholesteric lyotropic LC mesophase and eventually self-assembled into a solid sphere with average diameter of 3.4 $\mu$m, showing twisted-bipolar character. Such photoluminescent particles featured CPL with $|\delta_{\text{lum}}|$ of 0.23 in the suspension and exhibited remarkable angular anisotropy of $|\delta_{\text{lum}}|$ in the equatorial direction, 2.5-fold higher than that along the bipolar axis.
Moreover, Cheng et al. reported the first case of enhanced CPL materials prepared by adding LLC polymers to CLCs to form polymer-dispersed CLCs (PD-CLCs) (Figure 6).[87] They prepared binary CLCs by doping chiral AIE molecules (guest-1) into nematic LC 5CB as a host. The binary CLC exhibited weak CPL signals at approximately 615 nm with a maximum $g_{\text{lum}}$ of $+0.16/0.17$ for R/S-AD. Then, the new ternary achiral LLC polymer (LC-PPE, guest-2) was introduced into the above binary CLC to form PD-CLCs. These PD-CLCs showed significantly enhanced CPL centered at 447 nm, which belonged to LC-PPE. Notably, a very weak CPL signal of R/S-AD could still be detected. By regulating the concentrations of R/S-AD and LC-PPE, $g_{\text{lum}}$ values of CPL signals up to $+0.97/0.92$ could be achieved.

In summary, LLC-based CPL-active materials can be divided into three categories: (1) cholesterol derivatives with chiral moieties or $\pi$-conjugated polymers containing chiral side chains spontaneously forming a helical arrangement that generates a CPL signal; (2) achiral-conjugated polymers self-organized into a twisted structure under the induction of chiral dopants, some of which can further form higher-order assemblies driven by intermolecular interactions; and (3) LLC polymers introduced into small-molecular CLCs as templates to prepare polymer-dispersed CLCs that distinctly enhance the CPL emission. These CPL-emitting systems constructed with LLCs are capable of solving the incompatibility and instability issues of CLCs based on small molecules and feature outstanding processability and film-forming properties, which can broaden the applications of CPL materials in the solid state. However, the $g_{\text{lum}}$ values of CPL-active materials bearing LLCs are commonly in the range from $10^{-2}$ to $10^{-1}$ and have rarely been reported to reach 1.0. Thus, developing LLC-based CPL materials with large $g_{\text{lum}}$ values remains a formidable challenge.

**Organic–inorganic hybridized emitters**

In addition to pure organic emitters, some inorganic nanomaterials and organic–inorganic hybridized molecules can exhibit bright and intense fluorescence under excitation. Over the past years, chiral emissive materials fabricated by CDots, UCNPs and PKNCs have been developed.[88–90] Furthermore, metal–organic compounds containing rare-earth elements have enabled the achievement of upconverted CPL (UC-CPL).[91–94] This section will focus on the CPL emission originating from organic–inorganic composite systems, which can be classified into metal–organic complexes, CDots, and semiconductor NCs based on the emitter type.

**Metal–organic complexes**

Duan et al. provided the first example of enhanced UP-CPL in CLCs (Figure 7).[95] Blending a chiral emitter R/S-1 with a nematic LC (SLC-1717), a 10$^3$-fold amplified CPL was observed in CLCs compared with R/S-1 in solution. Subsequently, by adding an appropriate amount of Pt(II)octaethyl porphyrin as sensitizer, the UC-CPL of an anti-Stokes wavelength shift was obtained under 532 nm excitation through the triplet–triplet annihilation-based photon UC process. For comparison with the dilute solution, $g_{\text{lum}}$ was amplified by one order of magnitude to 0.04.

**CDots**

CDots are compatible with various nanoparticles and are ideal hosts for preparing CPL-emitting materials with the use of CDots. Xu et al. reported three-colored CDot nanomaterials based on CNCs that enabled strong, right-handed CPL with extraordinary $g_{\text{lum}}$ values of up to $-0.74$ (Figure 8A).[8]
The helical superstructures of CNC–CDot assemblies were induced by evaporation in homogeneous suspensions containing CNC and CDots. Three CNC–CDot films were obtained using a stoichiometric amount of CNC assembled with blue, green, and red CDots, denoted as CNC–B-CDots, CNC–G-CDots, and CNC–R-CDots, respectively, where the PBG bands were consistent with the emission peaks. Multicolor tunable CPL was realized through the choice of CDots and the PBG change. The PBG bands of the CNC–CDot films could be tuned from near-UV to near-infrared (NIR) by salt addition and ultrasonic treatment. When the emission band of CNC–B-CDots was superimposed on the PBG, strong right-handed CPL with $g_{\text{lum}}$ of $-0.74$ was detected (blue line); otherwise, the CNC–B-CDots exhibited weak CPL with $g_{\text{lum}}$ of $-0.045$ (black line). Furthermore, the influence of the fluorescence intensity on the CPL strength was investigated by changing the excitation wavelength. The results indicated that higher emission intensity led to stronger right-handed CPL accompanied by a change in $g_{\text{lum}}$ from $-0.24$ to $-0.32$.

Right-handed CPL based on CNC self-assembled helical superstructures has been extensively studied to date; however, the achievement of CP room-temperature phosphorescence (CPRTP) remains a considerable challenge owing to the instability of triplet-state excitons.[96,97] In their study, Liu et al. developed chiral photonic films that exhibited both CPL and CPRTP, which were constructed with CNCs, poly(vinyl alcohol) (PVA), and CDots (Figure 8B).[98] The self-assembly of CNCs provided chirality for CPL generation, and PVA was used to regulate the PBG of CNC/PVA hybrid films, leading to a tunable wavelength and invertible handedness of CPL, whereas CDots produced stable triplet excitons in this chiral photonic crystal environment. The CPL wavelength was red-shifted from 460 to 550 nm with increasing PVA content owing to the PBG change of the photonic films. It is worth noting that a positive CPL signal at approximately 480 nm was observed for the P3-CDot film, in addition to the weak left-handed CPL at 580 nm. Such left-handed CPL could be generated by the chiral arrangement of CDots using a CNC/PVA coassembly as a template. On the other hand, the emission peak of CDots was far away from the PBG band, resulting in a weak PBG effect. Consequently, invertible CPL was achieved by different PVA concentrations. PVA also restricted molecular motion and prevented oxygen quenching to protect triplet excitons. Moreover, by forming hydrogen bonds between the PVA and CDots, non-radiative relaxation could be effectively inhibited. Consequently, right-handed CPRTP with long lifetimes of up to 103 ms and a large RTP dissymmetric factor ($g_{\text{RTP}}$) of $-0.47$ was realized.

**Semiconductor NCs**

Semiconductor NCs have attracted considerable attention because of their high brightness, enhanced fluorescence quantum yields, good environmental stability, and narrow spectral emission. Extensive research on linearly polarized emission induced by NCs has been reported thus far, whereas CPL requires a helical arrangement, which seems more difficult. The use of CLCs as selective filters can transform the unpolarized luminescence from the emissive layer into high performing CPL, which can be modulated easily by shifting the overlap of the reflection and emission bands. Li et al. demonstrated a simple device that exhibited full-colored CPL accompanied by an extraordinary $g_{\text{lum}}$ of up to 1.6 (Figure 9A).[99] This device consisted of CsPbX$_3$ PKNCs, and two CLC polymer layers with opposite well-defined handedness. By varying the halide composition and regulating the nanoparticle size, inorganic CsPbX$_3$ PKs with blue,
green, and red emissions were obtained. The NC-containing film acted as an emissive layer. When the L-CLC and R-CLC films were placed on both sides of the PK film, fluorescence with the same handedness was reflected because of the selective reflection of CLCs, whereas the other half emission with the opposite helical sense could pass through. The unpolarized luminescence of PKNCs was fully converted into chiral emission, indicating that this proposed strategy enabled 100% circular polarization conversion efficiency. The handedness of CPL was confirmed using a polarizing filter.

Sun et al. demonstrated a CPL-active solid film by doping semiconductor CdSe/CdS QRs with red emission into the left-handed helical structure of CNCs (Figure 9B). The cationic QRs reduced the electrostatic repulsion between the anionic CNCs, which facilitated the self-assembly of the CNCs. The iridescent colors of the CNC-QR films changed from UV to red as the sonication time increased from 40 min to 90 min, whereas the red fluorescence of these films remained unchanged as the QR loading was constant. By adjusting the overlap area between the emission spectrum and...
the PBG band of the CNC–QR films, different CPL-active materials were realized, where $g_{\text{lum}}$ could reach $-0.45$. This work successfully extended self-assembled CP luminescent materials from fluorescent dyes and CDots to semiconductor NCs.

Benefiting from their excellent luminescent efficiency, CPL materials based on inorganic compounds exhibit large $g_{\text{lum}}$ values. There are three methods for preparing CPL materials: (1) doping of organic–inorganic hybridized emitters into CLCs, (2) coassembly of inorganic nanoparticles with CNCs, and (3) inorganic materials acting alone as luminescent sources, taking advantage of the selective reflection of CLCs to obtain high-performance CPL. Owing to the restrictions of the compatibility of inorganic nanoparticles in the host, CPL generated by inorganic-based systems has been relatively uncommon. However, some special CPLs, such as
UC-CPL, can only be formed by organic–inorganic materials, such as UCNPs, lanthanide-based metal-organic complexes, and chiral PK NCs. Therefore, considerable exploration remains to be conducted in this field.

### CPL REGULATION

The previous section reviewed several methods of CPL generation according to the emitter type, including the use of organic fluorescent molecules, LLCs, and organic–inorganic hybridized materials. To date, in addition to devoting themselves to the development of various CPL materials, scientists have shown substantial interest in tunable CPL-active systems that respond to external stimuli. This section will mainly focus on advanced and multifunctional CPL materials that respond to light, electricity, temperature, humidity, pH, and other factors.

#### Light irradiation

In recent years, photo-responsive soft materials have become a focus because of their non-contact, non-intrusive, and environmentally friendly properties. The photo-tunability of CPL, including the intensity and handedness, mainly depends on the photophysical and photochemical reactions of the emitters, such as Z/E photoisomerization and photocyclization, which can further affect the chiroptical properties of the chiral assembly. Our group has reported several photosensitive CLCs based on chiral fluorescence photoswitches. These photoswitches can undergo Z/E isomerization upon light irradiation, where the helical twisted power (HTP) value and fluorescence intensity vary between photostationary states (PSSs). For instance, by doping visible-light-driven chiral fluorescent molecular switch 4 into a nematic LC, a cholesteric phase was efficiently induced, which enabled chiral fluorescent molecular switch 4 into a nematic LC, ary states (PSSs). For instance, by doping visible-light-driven value and fluorescence intensity vary between photostationary values.

These photoswitches can undergo Z/E isomerization upon light irradiation, where the helical twisted power (HTP) value and fluorescence intensity vary between photostationary states (PSSs). For instance, by doping visible-light-driven chiral fluorescent molecular switch 4 into a nematic LC, a cholesteric phase was efficiently induced, which enabled strong CPL emission at 581 nm (Figure 10A). When irradiated with green and violet light, switch 4 showed large variations in both fluorescence intensity and HTP value, owing to the reversible trans-cis photoisomerization. Accordingly, the CPL signal of the switch 4-doped CLCs could be reversibly modulated by light irradiation at 520 nm and 405 nm.

Moreover, we proposed NIR-light-triggered reversible CPL modulation through the cooperation of a chiral fluorescence switch and UCNPs in CLCs (Figure 10B). The luminescent self-organized helical superstructure was constructed by doping switch 6 and UCNPs into a nematic LC. The NaYF₄-based UCNPs with core-shell-shell nanostructures exhibited different luminescent behaviors; the green emission was obviously enhanced. The PKNC emission was located at the center of the PBG. The UC-CPL could switch due to the reflection of the PBG. The UC-CPL emission and RET processes could be recovered by applying a mechanical force.

A photo-invertible helical cholesteric superstructure was designed using a chiral fluorescence photoswitch with photo-responsive properties (Figure 10C). The cholesteric helix was self-assembled by introducing left-handed switch 5 and a static dopant (CD-1) with opposite handedness into SLC1717. Consequently, the overall chirality originates from the contributions of both switch 5 and CD-1. Upon irradiation with 450 nm blue light, the HTP value of switch 5 decreased significantly, leading to the fast transition of the helical superstructure from left- to right-handedness, passing through the intermediate nematic phase. The helical sense of the CPL could be reversibly inverted between PSS₄₅₀ and PSS₃₆₅. The initial $g_{\text{lum}}$ of the right-handed CPL reached $0.55$.

#### Electric field

The electrical modulation of CPL is based on the electric field response of CLCs, which are well-known electric-optical devices that can freely switch between planar, focal conic, and homeotropic states when appropriate voltages are applied. The highly ordered helical supramolecular structure of CLCs can effectively induce strong CPL, whereas external electric stimuli disturb the arrangement of LCs, leading to a decrease or even disappearance of the CPL signal. Goto prepared a CPL-emitting device with a quick response to an electric field by employing phenylenevinylene-based chiral fluorescent dyes and 5CB. The CLCs showed intense CPL with a quite high $g_{\text{lum}}$ value of 0.6, and no CPL emission was observed under the application of voltage due to the cholesteric–nematic phase transition.

Cheng et al. reported a kind of “on-off-on” mode of recyclable CPL in CLC composed of two pairs of chiral fluorescent enantiomers and E7 host (Figure 11A). The CPL signals induced by these CLCs were in response to the electric field, accompanied by an LC phase transition from the planar state to the focal conic state, then to the uniformly lying helical (ULH) state, and finally to the homeotropic state. When the applied voltage was removed, the “on-off-on” $g_{\text{lum}}$ was detected, because CLCs in the ULH state could relax back to the planar state. Notably, the homotopic alignment of LCs could not relax to the original planar state unless re-annealing was performed over the clear point. Most interestingly, the larger $g_{\text{lum}}$ of CLC-2 could reach $0.83/0.86$, owing to the smaller dihedral angle of chiral fluorescent molecules R/S-M2.

Subsequently, Duan et al. demonstrated a significantly enhanced UC-CPL system through the synergy of the radiative energy transfer (RET) process and selective reflection of CLCs; in addition, the CPL could be switched under an electric field (Figure 11B). The CLC mixture was constructed by doping chiral dopant S/R-811, UCNPs, and CsPbBr₃ PKNCs into SLC-1717. By locating the emission of UCNPs at the left side of the photonic band edge, the UC luminescence was obviously enhanced. The PKNC emission was located at the center of the PBG. Benefiting from the RET process from UCNPs to PKNCs, the intensity of CPL emission from nematic LCs was not only undepressed, but also accompanied by an extremely high $g_{\text{lum}}$ of up to 1.1 due to the reflection of the PBG. The UC-CPL could switch under an applied electric field because the helical twist of CLC was unwound, and the LC molecules were arranged in the homeotropic alignment. Furthermore, the RET process exhibited an electrically controlled off-mode. Both the CPL emission and RET processes could be recovered by applying a mechanical force.
FIGURE 10 (A) Visible-light-driven circularly polarized luminescence (CPL) regulation through trans-cis photoisomerization of chiral fluorescence photoswitch. Reproduced with permission: Copyright 2019, Wiley-VCH. Ref. [108] (B) Near-infrared (NIR)-light-triggered CPL of self-organized emissive helical superstructures loaded with upconversion nanoparticles (UCNPs). Reproduced with permission: Copyright 2020, Royal Society Chemistry. Ref. [109] (C) Reversible chiral inversion of CPL in a photo-invertible helical superstructure induced by a chiral fluorescence photoswitch. Reproduced with permission: Copyright 2019, Royal Society Chemistry. Ref. [110]

Temperature

CLC-based emissive systems exhibit temperature-dependent CPL signals. When heated over the clear point of LCs, CPL can switch to the “off” state because the phase transition from the LC to isotropic state results in the disordering arrangement of LC molecules. Meanwhile, the reflection band red-shifts as the temperature increases in the CLC domain, which causes a change in the CPL intensity.

Akagi et al. constructed a thermally chiral-switchable CPL composite system with very high $g_{\text{Lum}}$ values of 1.1–1.6 at low and high temperatures, based on a racemic fluorescent polymer (diPA) and a double-layered CLC cell with opposite handedness at each layer (Figure 12). The design principle of this device relies on the selective reflections of CLCs with different helical senses and the thermally shifted reflective band of CLCs. The room-temperature LC (PCH302 and PCH304) and high-temperature LC (PCH3E02 and PCH3E04) as well as (R)-/(S)-chiral dopants were mixed to prepare left- or right-handed CLCs. The first cell layer was filled with left-handed CLC, and the reflection band was coincident with the fluorescence spectrum of the luminescent polymer. The CLC of the opposite handedness was injected into the second cell layer, showing a reflection band located at a shorter wavelength than the diPA emission. At low temperatures, the selective reflection of the right-handed CLC overlapped well with the photoluminescence band, leading to high-quality left-handed CPL. In contrast, the right-handed CPL was obtained at high temperature owing to the red shift in the reflection band of the CLCs.
Humidity

PBG changes have been widely employed to modulate the wavelength and strength of the CPL from CNCs. In addition, humidity-responsive CPL materials based on CNCs can be fabricated through ingenious self-assembly designs. Liu et al. demonstrated tunable UC-CPL by the coassembly of CNCs, UCNPs, and glycerol (Figure 13).[117] Glycerol was used to tune the PBGs of the chiral photonic films, which yielded a tunable wavelength of UC-CPL in the range from 450 nm to 620 nm with a tailored $g_{\text{lum}}$ value. Interestingly, due to the strong hygroscopicity of glycerol, G1-U revealed humidity-responsive UC-CPL emission. The $g_{\text{lum}}$ value at 450 nm decreased from 0.156 to 0.033 with an increase in relative humidity (RH) from 33% to 85%, attributed to the humidity-induced bathochromic shifts of the PBG and the reduced helical sense at higher RH.

Mechanical force

Mechano-responsive materials with reversible luminescent color changes have been extensively studied, but mechanically induced CPL properties are rare. In this regard, Tsukruk et al. developed a novel bio-inorganic elastomer by mixing the chiral coassembly of semiconducting quantum nanorods (QNRs) and CNCs with polyurethane (PU), which featured a dynamic chiro-optical property (Figure 14).[118] The core-shell CdSe/CdS QNRs could be seamlessly coassembled with CNCs by utilizing identical geometrical dimensions and complementary surface chemistries and oriented along the left-handed helical axis of the CNCs. The as-fabricated CNC–QNR–PU film showed mechano-optical behavior when stretched up to 70% due to the mechanically triggered transition from chiral helical organization to unidirectional nematic morphology. Accordingly, the reversible mechanical
deformation caused a reversible switch from CPL to linearly polarized emission.

**Multiple stimuli**

Tsukruk et al. constructed bio-organic CNC-based films whose handedness was altered by light illumination or an acidic environment (Figure 15A). This chiral emissive biocomposite was fabricated by integrating phase-shifting organic dye, which underwent reversible isomerization from ring-closed spiropyran (SP) to ring-open merocyanine (MC) upon light irradiation or the protonation of MC to protonated MCH$^+$ under acidic conditions, into CNCs through host-guest coassembly. The UV-treated CNC-SP film with the PBG band located at 645 nm showed strong right-handed CPL with $g_{\text{lum}}$ of $-0.4$, because the PBG band of CNCs well matched the emission peak of MCH$^+$ isomers at approximately 640 nm. After drying at 60°C or under water vapor treatment, the chemical conversion from MCH$^+$ to SPH$^+$ generated green emission at 535 nm due to reversible proton dissociation, giving rise to intense left-handed CPL with
FIGURE 14  Reversible switch from circularly polarized luminescence (CPL) to linearly polarized emission by mechanically triggered transition in the cellulose nanocrystal (CNC)–quantum nanorod (QNR)–polyurethane (PU) elastomer. Reproduced with permission: Copyright 2021, Wiley-VCH. Ref. [118]

A $g_{\text{ lum }}$ value as high as 0.61. In this case, the PBG of the chiral photonic film was not affected by the chemical stimuli-triggered isomerization, so the induction of left-handed CPL was ascribed to the inherent left-handedness of the CNC helical superstructures.

Deng et al. proposed a strategy that could be utilized to vary both the intensity and wavelength of CPL emission by controlling the fluorescence and chirality of CNC-based materials (Figure 15B).\[120] The composite films contained partially desulfurated CNCs and achiral fluorescent polymers with dansyl groups (PD) that could reversibly tune fluorescence in response to an acid or a base. The emitting-quenching mode of PD enabled controllable and repeatable CPL in the emission intensity of the PD/CNC films. Meanwhile, the helical pitch of the coassembly responded to the water content and polar solvent, causing a tunable PBG throughout the wavelength range of visible light. PBG changes further resulted in the wavelength-modulation of CPL. “On-off” CPL switching could also be realized by adding HCl or NH$_3$ to PD/CNC films with different water contents, indicating that the composite films showed responsiveness to multiple external stimuli simultaneously.

The co-stimulus-responsive behaviors of emitters and coassembly account for the CPL regulation of the entire system. The properties of CPL emitters remain unchanged in most CPL-tuning systems, whereas the coassembly superstructures are affected by stimuli; for instance, an increase in pitch can induce a red-shift of the PBG, and the unwound spiral structure can lead to vanished chirality. Another challenging modulation mechanism is that the chiroptical property of the chiral assembly changes under the inner driving force due to the physical or chemical reactions of emitters, such as photoisomerization or protonation. These stimulus-responsive CPL materials have great potential for the development of advanced information and display technologies.

CONCLUSION AND OUTLOOK

In recent years, CPL-active materials have attracted increasing interest and have been extensively studied owing to their wide application prospects. To date, several studies have demonstrated that the sources of CPL materials are extensive and involve many species such as lanthanide metal complexes, organic molecules, synthetic polymers, natural macromolecules, self-assembly systems, and chiral LCs. The development of CPL-active materials with high degrees of polarization has become urgent for actual applications in this field. Nevertheless, the $g_{\text{ lum }}$ values of most CPL materials do not meet the requirements of some optoelectronic applications. As mentioned above, a self-organized helical superstructure is an ideal candidate for constructing CPL-active materials with high $g_{\text{ lum }}$ and tunable properties. To date, scientists have achieved remarkable advances in CPL-active
materials by taking advantage of the cholesteric helical superstructure and other chiral LC materials. This review mainly recalled and systematically summarized the characteristics of CPL-active materials based on CLCs doped with different types of emitters including organic fluorescent molecules, LLCs, and organic–inorganic hybridized compounds. Among them, organic fluorescent molecules have better miscibility with LC molecules, and CPL with different emission band can be obtained by the flexible molecular design. Particularly, organic fluorophores with AIE property have higher fluorescence quantum yield, which exhibit stronger CPL than those common fluorescent dyes. Distinguishingly, chiral LLCs can engender CPL themselves. Organic–inorganic hybridized emitters are capable to induce the special UC-CPL and CPRTP; while the inorganic NCs featuring bright fluorescence and narrow emission are suitable for PBG-induced CPL. In addition, this review provided an overview of the dynamic switching of CPL signals in CLC-based CPL-active materials.

Although this review has highlighted the recent progress in CPL-active materials based on CLCs, many challenges remain in this rapidly growing research field. Specifically, there are three general directions for the future development of CPL-active materials based on CLCs. First, the original
mechanism causing the large dissymmetry values in CPL-active materials based on CLCs needs to be investigated and clarified in depth, which will provide key guidance for the design of next-generation CPL materials. Only when $g_{\text{lim}}$ is far beyond ±1, can the excellent performance of CPL-active materials be maximized in optoelectronic and photochemical applications. In fact, amplifying materials be maximized in optoelectronic and photochemical applications. This work provides valuable insight into the actual values due to the well-arranged helical superstructures of the design of next-generation CPL materials. Only when clarified in depth, which will provide key guidance for the active materials based on CLCs needs to be investigated and 20 of 23

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This review is expected to provide inspiration for follow-up investigations that can offer more open design perspectives for efficient CPL in CLC-based systems.

CONFLICT OF INTEREST

The authors declare that there is no conflict of interest that could be perceived as prejudicing the impartiality of the research reported.

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