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Advances in circularly polarized luminescence materials based on helical polymers

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Helical polymers are widely used in chiral recognition, chiral response, asymmetric catalysis, photoelectric functional materials, and other fields due to their unique helical structure and optical activity. In recent years, chiral helical structure and excellent processing properties have made helical polymers a new favorite in the field of circularly polarized luminescence (CPL) materials. In this review, CPL materials are briefly introduced, and then the advantages and classification of CPL materials based on helical polymers are discussed. According to the classification, the research progress of CPL materials of helical polymers is summarized. It is hoped that this brief review can stimulate the interest in helical polymers and promote the further development of helical polymers in the field of CPL.

Introduction

Helical structures widely exist in nature, such as DNAs and proteins in the microscopic world and the cosmic galaxies in the macroscopic world.[1,2] The helical structure is chiral, and the left-handed and right-handed helices are mirror images of each other. Inspired by this, people began to explore the ways of artificial synthetic helical polymers in the polymer material field. In the past decades, a variety of helical polymers have been designed and developed, including polyamides (PAIs), polyisonitriles (PINs), and polyacetylenes (PAEs). Due to their unique helical structures, these polymers can be widely used in chiral recognition, asymmetric catalysis, chiral separation, photoelectric functional materials, and other fields.[3–9]

Circularly polarized luminescence (CPL) has gradually become a new research hotspot in the photoelectric functional material field because of its potential in applications of various new areas, such as display technology, data storage processing, spin information communication, and other fields.[10–12] CPL refers to the phenomenon of left or right circular polarized light emitted by the chiral polarized materials, which can intuitively reflect the excited state structural information of chiral luminescence materials.[13–15] Fluorescence quantum yield (Φ_F) and dissymmetry factor (g_{lum}) are two important parameters for characterizing the properties of CPL materials.[16–20] The Φ_F refers to the ratio of the emitted photon number to the absorbed photon number, which is used to describe the emitting light ability of the material. g_{lum} refers to the difference ratio of left- and right-handed circularly polarized light to the total average luminous intensity, which characterizes the luminous intensity of the material. In earlier studies, the research works of CPL materials mainly focused on chiral transition metal complexes. The research found that the chiral lanthanide CPL complexes have high asymmetry factors, but their quantum efficiencies are generally low due to the existence of heavy atom effects.[21–23] In recent years, chiral organic small molecules and polymers have gradually become the new choices for CPL materials due to their advantages of easy modification and high quantum efficiency.[24–26] Although small organic molecules have high quantum efficiency, their g_{lum} values have been low. Compared with the CPL of small molecules, the CPL polymers can always exhibit a higher quantum efficiency and dissymmetry factor. The reason is CPL polymers possess higher molecular weight and more repetitive units than the CPL molecules, which can amplify the luminescence properties and chiral signals.[28–30] In addition, CPL polymers also have excellent photoelectric properties and good processability. As a result, CPL polymers have attracted more and more attention in recent years.

Helical polymers with CPL activity (CPL helical polymers, CHPs) combine chirality and chromophores. These two outstanding characteristics of CHPs make it a classical CPL material. According to the construction ways, CHPs can be divided into four categories:

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(1) CPL based on optically active helical polymers: CHPs are obtained from CPL small molecules by homo-polymerization or copolymerization and using covalent bonds for chiral transfer. (2) CPL based on chiral induced helical polymers: under the condition of external chiral environment induction, a chiral transfer is carried out through non-covalent interactions to obtain chiral helical polymer containing chromophore. (3) CPL based on self-assembled helical polymers: in the absence of an external chiral environment, molecules self-assemble through non-covalent interactions as the driving force to form stable chiral helical conformation and obtain CPL ability. (4) CPL based on composite helical polymers: no external chiral environment and any covalent bonds or non-covalent bonds, the chiral unit and fluorescent unit are directly combined, and then, the chiral structure of the helical polymer is used to select and absorb the light emitted from fluorescence source to obtain helical polymer composite material with CPL ability.

CPL based on optically active helical polymers

The most direct and effective way to construct CPL materials is to transfer chirality through covalent bonds by homopolymerization or copolymerization to obtain chiral helical macromolecules containing chromophores.[13–31]

According to the report, the first CHP is PDMBT (Fig. 1), a polythiophene derivative based on chiral side chain modification with g_{lum} up to $(5.0 \pm 1.0) \times 10^{-3}$. It was developed by Meijer *et al.* in 1996.[32]

In 2013, Cheng *et al.* designed and synthesized a chiral copolymer TPETyr (Fig. 1) containing TPE units by Sonogashira coupling reaction, which exhibited a high asymmetry factor in THF solution ($g_{\mathrm{lum}} = 0.44$, $\Phi_F = 3.6\%$). (Fig. 2).[33] Two years later, the CPL copolymer R/S-P (Fig. 1) was reported by the same team, and it was prepared through copolymerization of luminescent chromophore phenothiazine and chiral 1,1′-naphthalenyl-2,2′-diol (BINOL). The test results show that its g_{lum} can achieve 1×10^{-3}.[34] Based on these works, Cheng's group further synthesized a class of four-component helical polymers in 2017, and a deep red CPL signal (g_{lum} up to 2×10^{-3}) can be tested due to intramolecular Förster resonance energy transfer (FRET).[35] Subsequently, a series of related research works were expanded by this team for demonstrating that dinaphthalene based chiral polymers are one of the best choices for constructing CHPs.[36–40]

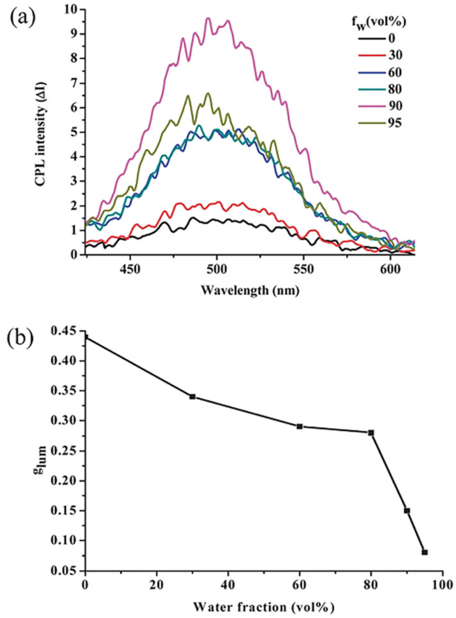

Fig. 2 (a) CPL spectra of TPETyr in THF/water mixtures with different fractions of water; (b) CPL dissymmetry factor g_{lum} *versus* water fraction for TPETyr. The g_{lum} (508 nm) = 0.44, and the Φ_F = 3.6%. Copyright 2013, Royal Society of Chemistry.

In 2014, a quinoline-based helical polymer with chiral side chains was prepared by Suginome *et al.* It was found that the CPL reversed with the replacement of the solvent from trichloromethane to 1,1,1-trichloroethane. They attributed this finding to the change of the *M*-helix to the *P*-helix structure.[41] Subsequently, they also developed quinoline-based copolymers containing different copolymerization units and achieved colour modulation. These works demonstrate the potential application of such helical polymers in the CPL field.[42,43]

In 2016, inspired by the one-hand excess helical structure of ellagitannin, Ikai *et al.* designed and synthesized optically active polythiophene (poly-9, Fig. 1). It is worth noting that poly-9 shows a strong CPL emission property ($g_{\mathrm{lum}} = 1.9 \times 10^{-2}$, $\Phi_F = 13\%$) because the biphenyl structure units linked single-handed excess axial deformation glucose units are introduced into its backbone.[44] In 2017, they synthesized a series of cellulose-derived polymers that also possess CPL activity and have a g_{lum} of 3.0×10^{-3}.[45] Subsequently, a series of chiral polymers with glucose groups, also possessing CPL activity, was developed on the basis of these works.[46–49] These results show that chiral biomass resources have the potential to be developed into novel CHPs materials.

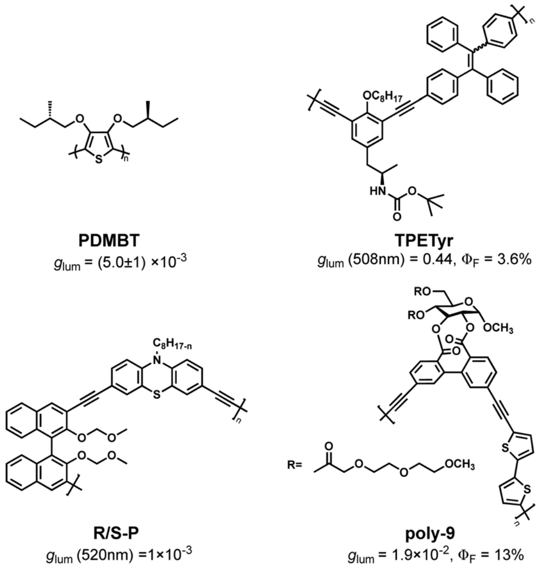

Fig. 1 Chemical structures of optically active helical polymers.

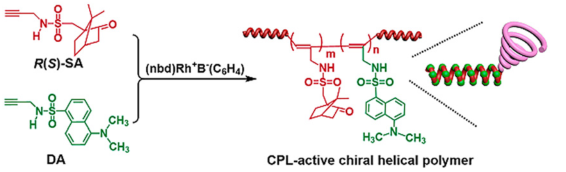

Fig. 3 Schematic illustration for preparing fluorescent chiral helical monosubstituted polyacetylenes with CPL performance. Copyright 2018, American Chemical Society.

In 2018, a CPL-active chiral helical polymer was (Fig. 3) reported by Deng *et al.*[50] This polymer was prepared by copolymerizing achiral acetylenic fluorescent unit and chiral unit and exhibited excellent optical activity and CPL capability with g_{lum} of 10^{-1} (Fig. 4). The single helical structure R(*S*)-SA of the copolymer plays a dual role in achieving the CPL process: one provides helical chirality for the CPL materials, and the other effectively suppresses the aggregation-caused quenching (ACQ) effect of the fluorescent groups. Subsequently, in 2020, Deng *et al.* prepared a CPL copolymer consisting of chiral units and achiral alkyne units containing tetraphenylene (TPE) and found that the polymer can exhibit a high g_{lum} value of CPL emission in solid films, while no CPL emission was detected in the dispersed state.[51]

Wan *et al.* reported the first example of luminescent monosubstituted polyacetylenes (mono-PAs) based on a contracted *cis*-cisoid polyene backbone. It has an excellent CPL performance and has a g_{lum} of 10^{-1}.[52] They developed a series of mono-PAs that can recognize structurally diverse achiral amines. The achievement of this function relies on the *cis*-transoid to the *cis*-cisoid helical transition of the polyphenylacetylene backbone.[53]

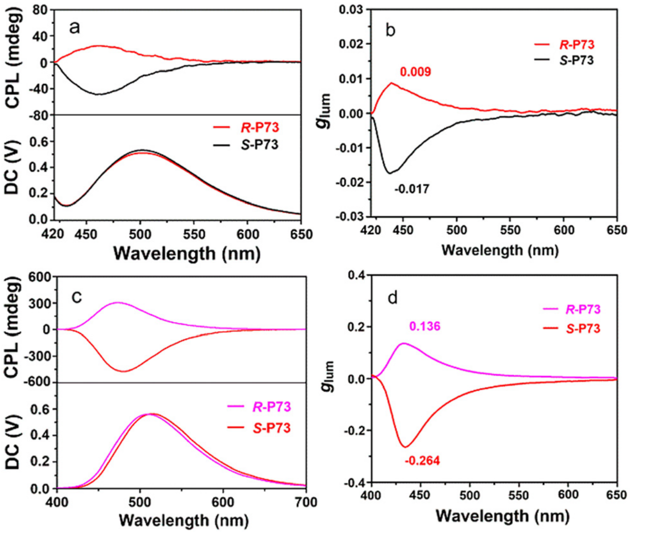

Fig. 4 (a and c) CPL spectra of R-P73 and S-P73 in $CHCl_3$ solution excited at 369 nm. (b and d) CPL dissymmetry factor g_{lum} *versus* wavelength of R-P73 and S-P73 excited at 369 nm. In (a) and (b), [R-P73] = [S-P73] = 1 mM; in (c) and (d), [R-P73] = [S-P73] = 10 mM. The g_{lum} (R-P73 in 435 nm) = 0.136, and the Φ_F = 25.3%. The g_{lum} (S-P73 in 435 nm) = −0.264, and the Φ_F = 17.6%. Copyright 2018, American Chemical Society.

In 2021, using a chiral donor–acceptor copolymerization strategy, Chen *et al.* developed a series of chiral non-conjugated polymers (R, R)-/(S, S)-pTpAcDPS, (R, R)-/(S, S)-pTpAcBP, S–P and R–P with excellent circularly polarized electroluminescence (CP-EL) capability and have a g_{EL} of 10^{-3}.[54,55] Cheng *et al.* introduced a chiral functionalized bis-benzoxanethonyl unit by intramolecular chiral transfer mechanism to prepare three conjugated polymer backbones (S-/R-BP, S-/R-WP1, and S-/R-WP2) to construct full-colour CP-EL.[56]

In addition, Chen *et al.* synthesized a pair of linear axially chiral conjugated polymers by incorporating rigid axially chiral chromophores into the polymer backbones.[57] The twisted biphenyl skeleton effectively reduces the energy gap between the S1 and T, which greatly facilitates the intersystem crossing (ISC) process. The unique structural design makes it have low-temperature UOP (ultralong organic phosphorescence) and CPL properties.

In general, the construction of intrinsic CPL-helical polymers by homopolymerization and copolymerization is one of the most common and simplest methods.

CPL based on chiral induced helical polymers

In fact, the chiral transfer not only can be achieved by the covalent bonds but also by the non-covalent bonds when some polymers in a chiral environment, such as chiral solvents, small chiral molecules, and circularly polarized light.[58–64] Therefore, the chiral structural character can be transferred to polymers' chromophores by non-covalent interactions under a chiral environment; as a result, the polymers can exhibit CPL ability.

In 2010, Fujiki *et al.* found that the achiral conjugated polymer PF10 can produce asymmetric aggregation in a chiral mixed solvent. The chiral solvent consists of a large amount of R/S-limonene (the main solvent), a small amount of chloroform (the good solvent), and a small amount of methanol (the poor solvent) (Fig. 5).[65] Under this chiral environment, the backbone of polymer PF10 was arranged in a single helical direction to realize the chiral transfer successfully so that PF10 not only acquired chirality but also possessed CPL ability (g_{lum} = $\pm 2.33 \times 10^{-3}$, Φ_F = 59%). In subsequent studies, the chiral transfer of more achiral conjugated polymers in chiral solvents was explored, and these research results laid a foundation for further exploring solvent-induced CPL-helical polymer materials.[66–69]

In 2012, Akagi *et al.* used liquid crystalline polyacetylene (diLCPA) derivatives bearing 4-nonyloxy phenyl groups and chiral dopants to obtain a chiral nematic liquid crystal material with excellent CPL capability with an asymmetry factor of 10^{-1}.[70]

Using the chiral solvent introduction method, some achiral branched polymers were also investigated by Zhang *et al.* By the chiral solvent introduction, a series of achiral polymers with azo side chains and achiral polyfluorene polymers successfully obtained the CPL activity. These results further enriched the

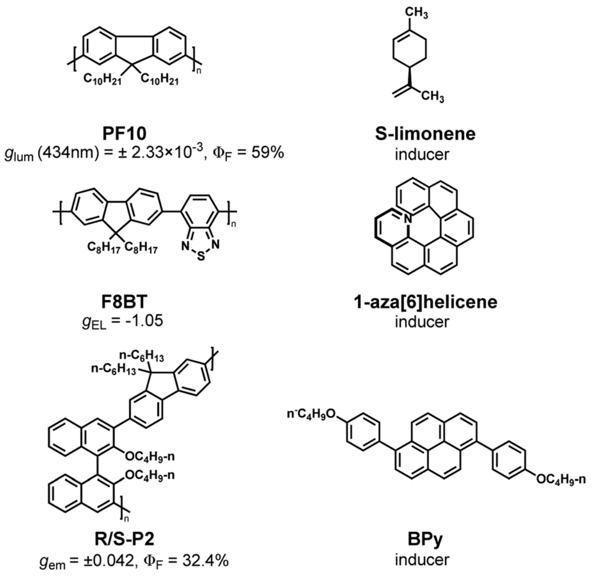

Fig. 5 Chemical structures of chiral induced helical polymers.

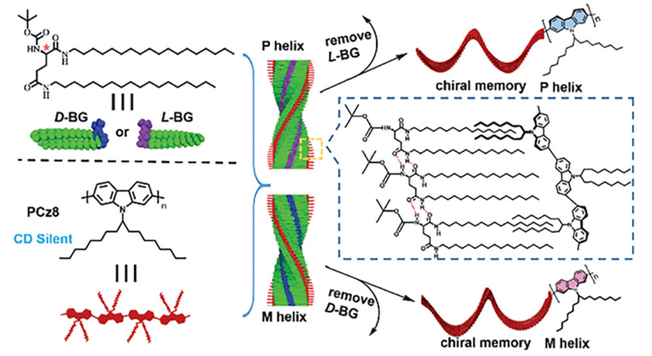

Fig. 7 Illustration of chiral transfer from gelator BG to achiral polymer and chiral memory effect. Copyright 2016, Royal Society of Chemistry.

research work on solvent-induced CPL-helical polymer materials.[61–76] In addition, they also proposed a co-gel method for realizing the chiral induction based on achiral polymer PCz8 and chiral D/L-BG gels.[77] In the mixed system, the gel agent BG is the chiral matrix material, which can induce the polymer PCz8 to form a helical conformation through the entangling of alkyl chains during the formation of the co-gel. After removing the chiral gel matrix, the induced chirality of the polymer PCz8 can be maintained, resulting in a chiral helical polymer with CPL ability (Fig. 7).

Fuchter *et al.* doped the chiral aza[6]helicene molecules into the copolymer of dioctylfluorene and benzothiadiazole (F8BT).[78,79] Under the π–π interaction, the backbone of polymer F8BT was induced to form a helical conformation, which made the composite system F8BT/aza exhibit obvious CPL characteristics with g_{EL} = −1.05 (Fig. 5). In 2020, Cheng *et al.* used another chiral doped small molecule, R-/S-3, to induce F8BT polymer and also obtained the helical polymer with CPL characteristics.[80] Fuchter *et al.* and Ji-Seon Kim *et al.* successfully prepared materials with CP-EL capability using F8T2 and F8BT after chiral aza[6]helicene molecule induction. They have a g_{EL} of −0.65 and 0.3, respectively.[81,82] These results indicate that F8BT with chiral helical structure has great application potential in CP-OLED.[78–80,82]

In 2020, Zou *et al.* found that after the irradiation of left-handed or right-handed CPL light, the CPL signal can be measured from achiral luminescent polymer F6BT.[83] The intensity and direction of the generating CPL signal are strongly influenced by the polarization direction and irradiation wavelength of the chiral source CPL light.

In 2021, Cheng *et al.* used the chiral non-fluorescent polymer R/S-P (1-3) to induce small fluorescent molecule BPy through π–π interaction during thermal annealing, and this system also showed obvious CPL characteristics and had a g_{em} of ±0.042 (Φ_F = 32.4%) (Fig. 5 and 6).[84] In 2022, Cheng *et al.* used two chiral binaphthyl polymers as chiral inducers to construct co-assemblies with achiral pyrene-naphthalimide dye (NPy) and obtained an excellent CP-EL performance (g_{EL} = 4.8 × 10^{-2}).[85] Subsequently, they used three achiral conjugated pyrene-based dyes (BP, w-WP, and c-WP) doped with chiral binaphthyl-based enantiomers (S-/R-M) and successfully obtained nanofibers with regular helical structure by chiral co-assembly under the effect of thermal annealing.[86] This material has excellent CP-EL capability with a g_{EL} of 6.2 × 10^{-2}. In subsequent work, they chose three achiral liquid crystal polymers (LC-P1/P2/P3) and chiral dinaphthyl inducers

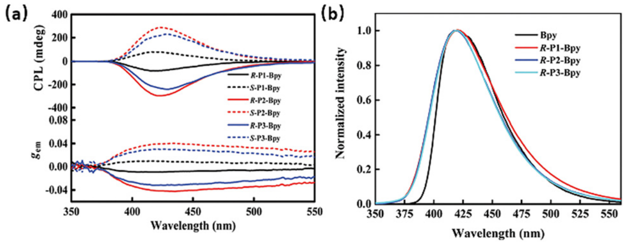

Fig. 6 (a) CPL spectra of R/S-P1-Bpy, R/S-P2-Bpy and R/S-P3-Bpy in spin-coated films after annealing. (b) PL emission of Bpy, R-P1-Bpy, R-P2-Bpy, and R-P3-Bpy in spin-coated films. The |g_{em}| (R/S-P2-Bpy in 420 nm) = 0.042, and the Φ_F (R-P2-Bpy) = 32.4%. Copyright 2021, Royal Society of Chemistry.

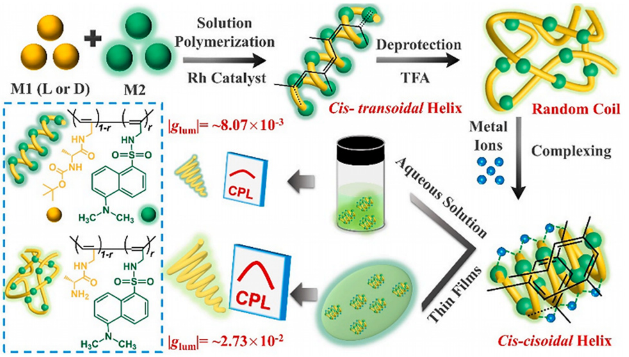

Fig. 8 Schematic illustration for the aqueous phase CPL-active materials with a *cis*-cisoid helical structure based on the random coil to one-handed helix transformation induced by metal coordination. Copyright 2022, Elsevier Ltd.

(R/S-M) with anchored dihedral angles to construct chiral co-assembled liquid crystal polymers.[87] It was found that regular helical nanofibers with CPL activity signals could be generated after thermal annealing.

In 2022, Li *et al.* exploited the synergistic effect of hydrogen bonds and metal coordination in an aqueous solution and successfully induced a single chiral helical polymer with CPL activity (Fig. 8).[88]

In the chiral environment, non-covalent interactions, such as molecular π–π interactions, hydrogen bonds, and chiral light sources, can be used to carry out the chiral transfer and obtain CPL active helical polymers, which is also a common method to obtain CPL-helical polymers.

CPL based on self-assembled helical polymers

In addition, only relying on the interaction between molecules themselves, the ordered helical chiral structure can also be obtained by self-assembly behaviour without a chiral environment. Therefore, the chiral polymer with CPL ability also can be obtained in this way.

In 2015, Cheng *et al.* synthesized a chiral conjugated polymer P-1 (Fig. 9) containing (R)-1,1′-binaphthyl and tetraphenylethene moieties. The CPL signal was not found in the THF solution of P-1 but was detected in its THF/H_2O mixed solution. That is because P-1 can induce self-assembly behaviour into helical nanofibers by π–π interactions in its THF/H_2O solution. The helical nanofibers of P-1 have CPL ability ($g_{\mathrm{lum}} = -1.6 \times 10^{-3}$).[89]

Tang *et al.* introduced TPE into the conjugated polymer backbone, and copolymer P(TPE-alanine) was obtained, which had excellent AIE and CPL performance ($g_{\mathrm{em}} = 0.0045$, $\Phi_{\mathrm{F}} = 16.8\%$).[90] More interestingly, the copolymer self-assembled in the THF/H_2O system. With the increase in water content, its self-assembled form changed from microspheres to fibres. The self-assembled polymers exhibited CD and CPL properties, and the g_{lum} reached 0.045. Subsequently, they constructed a polymer with a similar structure, P(TPE-phenylanine), and studied the self-assembly behaviour of P(TPE-phenylanine) under the condition of deprotection and adding chiral additives (Fig. 9).[91] With the improvement of hydrogen bonding, the self-assembly behaviour also changed, the helical nanofibers were gradually formed, and the corresponding CPL ability was gradually improved ($g_{\mathrm{em}} = 0.018$, $\Phi_{\mathrm{F}} = 20.7\%$) (Fig. 10).

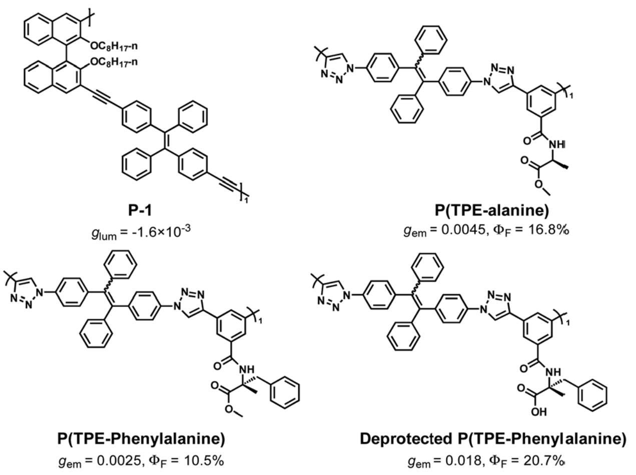

Fig. 9 Chemical structures of self-assembled helical polymers.

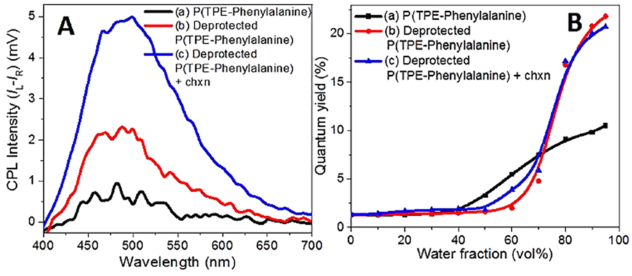

Fig. 10 (A) CPL of P(TPE-phenylalanine), deprotected P(TPE-phenylalanine), and deprotected P(TPE-phenylalanine) + chxn. (B) Variations in the fluorescence quantum yields of these three kinds of polymers in THF/water mixtures with different water fractions. The g_{lum} (deprotected P(TPE-phenylalanine) + chxn) = 0.018, and the Φ_{F} = 20.7%. Copyright 2020, American Chemical Society.

In 2020, our group designed and synthesized the P3HT-***b***-PPI block copolymer modified with chiral alkyl side chains.[92] Using the excellent crystallinity of P3HT and the chirality of PPI, this block copolymer can realize the crystallization-driven asymmetric self-assembly (CDSA) to form a single helical chiral fibre with controllable length, narrow dispersion, and clear handedness. In the process of self-assembly, the chiral property is transferred from PPI to supramolecular assembly, which makes the copolymer assembly have good chiral optical activity. The helical assembly formed by the chiral block copolymer exhibits white light emission and excellent CPL performance (Fig. 11). Then, we reported the controlled synthesis of π-conjugated poly(phenyl isocyanide)-*b*-poly(phenyleneethylene) (PPI-*b*-PPE) copolymers *via* chain extension of ethynyl 4-iodobenzene

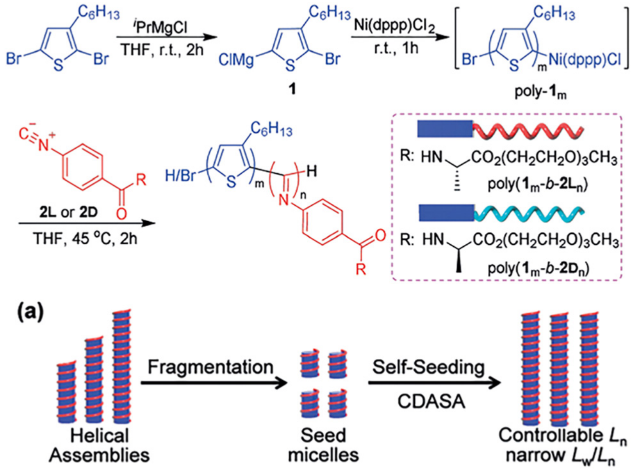

Fig. 11 Synthesis of P3HT-*b*-PPI copolymers(top). Schematic illustration of the preparation of narrow dispersed cylinder-like helical micelles *via* self-seeding method (down). Copyright 2020, WILEY-VCH.

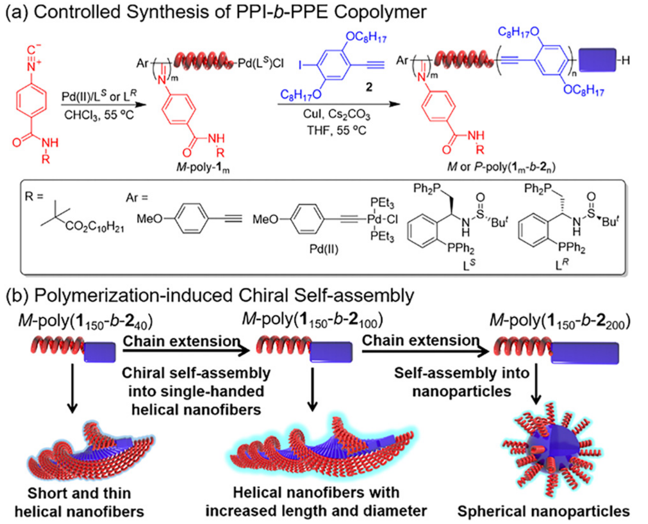

Fig. 12 (a) Controlled synthesis of the PPI-*b*-PPE copolymer; (b) schematic illumination of polymerization-Induced chiral self-assembly. Copyright 2021, American Chemical Society.

initiated by Pd(II)-terminated helical poly(phenyl isocyanide) (PPI).[93] The polymerization induced chiral self-assembly (PISA) of the resulting block copolymers, resulting in one-handed helical nanofibers with defined helicity and controllable size. Interestingly, the helical chirality of PPI is transferred to the supramolecular structure, resulting in high optical activity of the supramolecular nanostructure and the clear emission of CPL (Fig. 12).

Recently, we synthesized a series of copolymers, poly(cholesterol allene)-*b*-poly(3-hexylthiophene) (PCA-*b*-P3HT) containing helical PCA and poly(alkoxy allene)-*b*-poly(3-hexylthiophene) (PAA-*b*-P3HT) containing achiral PAA segments by using a nickel catalyst.[94] Crystallization of P3HT and helicity of PCA drove PCA-*b*-P3HT self-assemble into spherical nanoparticles that gradually transformed into one-handed helical nanofibers by CDSA. The chirality of PCA was transferred to the supramolecular structure, which induced high optical activity of P3HT. The chiral seed micelles of PCA-*b*-P3HT were found to induce asymmetric block copolymerization of achiral PAA-*b*-P3HT copolymers, which led to one-handed helical supramolecular block copolymers. Although PAA-*b*-P3HT is achiral, the block copolymerization proceeded in a helix selective manner, giving the supramolecular block copolymers controlled helicity. Consequently, the supramolecular block copolymers showed interesting white-light emission and CPL (Fig. 13).

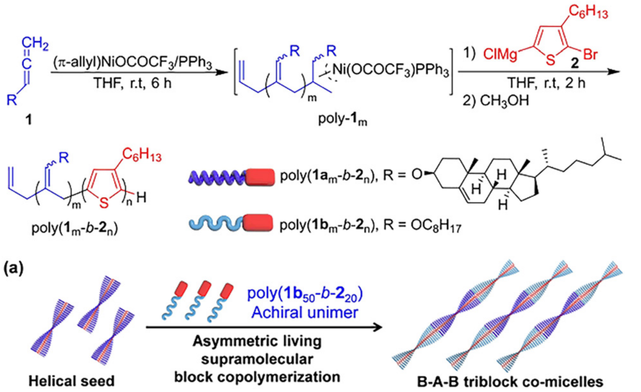

Fig. 13 Synthesis of P3HT block copolymers (top). Schematic illustration of asymmetric supramolecular polymerization of achiral poly($\mathbf{1b}_{50}$-*b*-$\mathbf{2}_{20}$) unlimber initiated by right-handed helical seed micelle of poly($\mathbf{1a}_{80}$-*b*-$\mathbf{2}_{20}$) (down). Copyright 2022, WILEY-VCH.

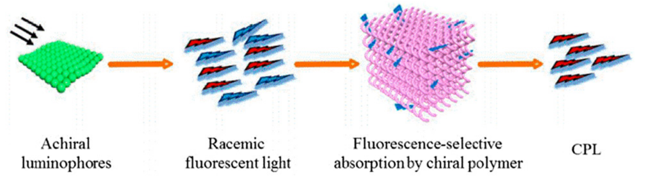

Fig. 14 Schematic illustration of the fluorescent selective absorption mechanism. Copyright 2019, American Chemical Society.

In summary, due to the complex structural design and control of self-assembly, few self-assembled CPL-helical polymer materials have been reported so far. It is hoped that this review will give researchers new enlightenment and research ideas.

CPL based on composite helical polymers

In addition to covalent and non-covalent bonds, a new method to construct CPL-helical polymer materials has emerged in recent years. The chiral polymer materials with CPL ability were obtained by combining the chiral polymer and fluorescent molecules and using the chiral structure of the chiral polymer to screen the light emitted by the fluorescence source.

Deng *et al.* established a strategy to prepare CPL-helical polymers based on chiral helical polyalkyne and achiral fluorescent units.[95,96] Based on this strategy, they proposed the ''matching rule'', which means that when the CD signal wavelength of the chiral helical polyalkyne partially matches the fluorescence emission wavelength of the fluorescence unit, the CPL signal can still be generated in the separated state even if they have no chemical interaction. This is mainly attributed to the helical polymer acting as a filter to screen the light emitted by the fluorescent source to obtain circularly polarized light with single handedness (Fig. 14). Based on this strategy, they constructed CPL materials of full colour and fabricated CPL devices with switching responsiveness using binary and ternary chiral fluorescence films, with a maximum g_{lum} of +0.323. Then, they constructed a composite of chiral helical polymers with perovskite materials, which also had CPL activity.[97] A series of CPL helical polymer composites with different colours were obtained by electrospinning.[98]

Conclusions and outlook

This review describes the research progress of CPL materials constructed by different methods based on helical polymers. Firstly, helical polymers and circularly polarized luminescence are briefly introduced. According to the different ways of constructing circularly polarized luminescence materials, helical polymers are divided into four categories. Then, according to these classifications, the research trends of helical polymers in the field of CPL in recent years are described. It can be found that researchers have developed various spiral polymers with CPL activity through different methods, but few of the CPL helical polymer materials can break the g_{lum} of 1. Therefore, we believe that the future development direction and difficulty of CPL-helical polymers lie in how to improve the asymmetry factor through reasonable structural design or other means, such as doping. Moreover, the mechanical properties and stability of the polymer material, as well as the availability of mass production, will be the main factors affecting the development of CPL-helical polymers if practical applications are considered. In conclusion, challenges and opportunities exist, and it can be expected that the exploration of CPL-helical polymer materials has great significance and broad application prospects.

Author contributions

S.-Y. L, L. X., and R.-T. G. wrote and edited the paper. Z. C., N. L., and Z.-Q. W. conceived the idea and corrected the manuscript.

Conflicts of interest

There are no conflicts to declare.

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