Through-space charge transfer polymers for solution-processed organic light-emitting diodes

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Abstract
Through-space charge transfer (TSCT) polymers are an attractive class of luminescent polymers with spatial donor/acceptor architecture and thermally activated delayed fluorescence effect, different from conventional luminescent polymers with conjugated donor-acceptor structure and through-bond charge transfer emission. Their emission comes from the intramolecular charge transfer by through-space pathway because the donor and acceptor segments are spatially proximate to each other in each repeating unit but are physically separated by nonconjugated polymer backbone. In this review, recent advances in TSCT polymers with linear, bottlebrush, and dendritic architectures are presented, with the focus on their molecular design, photophysical behavior, and device performance. We hope that this review shall provide a useful insight of new luminescent polymers with TSCT effect for use in solution-processed organic light-emitting diodes.

KEYWORDS
luminescent polymer, organic light-emitting diodes (OLEDs), solution process, through-space charge transfer

1 | INTRODUCTION

Charge transfer (CT) interactions between electron donors (D) and electron acceptors (A) are the basis of optoelectronic functions of many organic and polymer materials,[1–4] which have enabled important applications including organic light-emitting diodes (OLEDs),[5–7] chemical sensors,[8,9] and bioimaging.[10] In general, CT process can be divided into two kinds according to the interaction pattern between donor and acceptor. One is through-bond charge transfer (TBCT) mediated by conjugated bonds,[1] which features strong electron coupling between donor and acceptor through covalent bonds, while the other is through-space charge transfer (TSCT), which occurs through spatial electron interactions and has relatively weaker electron coupling between donor and acceptor.[11–25]

Since the discovery of electroluminescence from poly(p-phenylene vinylene) (PPV) in 1990,[26] solution-processed OLEDs based on luminescent polymers have attracted much attention because they are compatible with low-cost and large-scale processing technologies including spin coating and ink-jet printing.[27–29] According to the emission mechanism, electroluminescent polymers can be divided into three kinds: fluorescent polymers, phosphorescent polymers, and thermally activated delayed fluorescence (TADF) polymers. For fluorescent polymers, only singlet excitons can be utilized for light emission due to spin statistic rule, leading to maximum internal quantum efficiency (IQE) of 25%.[30] In contrast, phosphorescent polymers can harvest both singlet and triplet excitons through intersystem crossing (ISC) process of heavy metal complexes, enabling them to reach 100% IQEs.[31,32] Recently, TADF polymers have emerged as promising materials for solution-processed OLEDs because they can convert triplet excitons into singlet ones through reverse intersystem crossing (RISC) process[33,34] to realize 100% IQE without using noble metal complexes, becoming an important approach for the development of luminescent polymers.[35,36] Based on the nature of polymer backbone, there are two kinds of polymers: one is conjugated polymer in which the π-electron cloud can be delocalized along the main chain, the other is nonconjugated polymer backbones where the repeating units are electrically isolated from each other, and the emissions are coming from the attached emissive species.[37] So far, the majority of luminescent polymers with CT characters are based on conjugated D-A architectures and TBCT emissions.[1,5–7] The introduction of electron donor and acceptor segments into the conjugated polymer not

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only allows to tune the emission color by adjusting the donor and acceptor strength, but also can tune the energy levels of excited state to realize efficient exciton utilization. For example, polyfluorene\cite{38–42} and polyspirofluorene derivatives\cite{43,44} containing D-A structure with TBCT emission are proved to be a successful approach to realize full-color emission covering the whole visible region for solution-processed OLEDs. Recently, TADF polymers\cite{45–59} with finely controlled CT processes based on conjugated donor-acceptor architectures have been demonstrated to utilize nonemissive triplet excitons through RISC process. However, the strong through-bond charge transfer effect in conjugated architecture tends to induce a large redshift of emission, which is undesirable for realizing blue emission. Another challenging issue for the development of luminescent polymers with CT emission for TADF effect falls in the contradiction between small singlet-triplet energy splitting ($\Delta E_{ST}$) and high photoluminescence quantum yield (PLQY), that is, the sufficient separation of HOMO and LUMO required for small $\Delta E_{ST}$ could lead to small oscillator strength and thus low PLQY.\cite{60} Therefore, it is desirable to develop new approach for the balanced control of charge transfer strength, $\Delta E_{ST}$, and PLQY for designing luminescent polymers with tunable emission color and TADF effect.

Different from conventional luminescent polymers with through-bond charge transfer emission, our group proposed the new strategy to develop TSCT polymers, which consists of nonconjugated polymer backbone (polystyrene as backbone) and spatial donor/acceptor architectures for achieving TADF effect in 2017.\cite{61} In this motif, donor and acceptor segments are physically separated, but meanwhile are spatially proximate to each other, allowing the through-space, rather than through-bond charge transfer to occur. This molecular design has the following advantages. First, nonconjugated backbone can avoid strong electron coupling between donor and acceptor segments, favorable for realizing blue emission. Second, the physical separation of donor and acceptor can lead to small overlap of HOMO and LUMO distribution and thus small $\Delta E_{ST}$.\cite{11,12} Third, the electron clouds of donor and acceptor can communicate with each other through spatial interactions to enhance the radiative decay rate, giving promising PLQY for the designed polymers. Based on this strategy, three kinds of TSCT polymers with different topological structures, including linear,\cite{61–66} bottlebrush,\cite{67} and dendritic architectures\cite{68,69}, have been developed so far (Figure 1). In this review, current advances in TSCT polymers for solution-processed OLEDs are summarized. Their molecular design, optoelectronic property, structure-property relationship, and device performance are discussed. We hope that this review not only provides a comprehensive summary of TSCT polymers, but also offers molecular design guideline for the future development of luminescent polymers possessing novel optoelectronic properties.

2 | TSCT POLYMERS

2.1 | TSCT polymers with linear architecture

According to the polymer backbone, TSCT polymers with linear architectures can be divided into two kinds: polystyrene-based TSCT polymers and polynorbornene-based TSCT polymers. Polystyrene can be used as the backbone because the pendent phenyls are aligned.
parallelly with close distance of 2.55 Å in the extended conformation,[70] enabling intramolecular through-space π-π interaction between adjacent phenyls. Moreover, the nonconjugated structure of polystyrene backbone can avoid the large electron cloud delocalization between donor and acceptor segments, favorable for achieving blue emission. Based on the polystyrene backbone, TSCT polymers are designed by attaching acridan (Ac) as donor and triazine (TRZ) as acceptor in side chain through meta-phenyl linkages (P1, Figure 2).[61] The acridan and triazine units are selected because of their quasi-planar structures that are favorable for enhancing through space interaction, while the meta-phenyl linkages are used to keep their electronic properties unaffected by polymer backbone. To explore the effect of relative mole ratio between donor and acceptor segments on emission properties of TSCT polymers, the contents for acceptor segments are fixed at 5 and 50 mol%, respectively. Moreover, to investigate the nature for TSCT effect, control polymer containing four tert-butyl groups on each acridan unit (P2) are designed to investigate the steric hindrance effect on TSCT emission. Density functional theory calculation results show that HOMOs of the resulting polymers are localized on the acridan segment, whereas LUMOs are distributed on triazine, indicating CT character of these polymers. P1 shows broad and solvent-dependent emission bands with the peak red-shifted by 51-113 nm relative to the homopolymers containing only acridan (P-Ac) or triazine segments (P-TRZ) in side chain. Moreover, the separated frontier molecular orbitals result in small $\Delta E_{ST}$ of 0.019-0.021 eV, making them capable to convert triplet excitons to singlet ones by RISC process. TADF effect for P1 are supported by transient photoluminescence (PL) spectra, which display distinctive delayed fluorescence with lifetime ($\tau_d$) of 1173.0-1279.4 ns in addition to prompt fluorescence with lifetime in nanosecond scale. For P2 containing tert-butyl groups on donors, PL spectra exhibit emission from the individual donor and acceptor segments without CT emission, and the delayed fluorescence is not detected, indicating that CT interactions and TADF effect can be suppressed due to the steric hindrance. On another hand, the physical mixture of homopolymer P-Ac and P-TRZ mainly exhibit emissions from donor and acceptor segments, accompanied with much lower PLQY (13%) compared to copolymer P1 (60%). This result indicates that copolymerization of donor and acceptor in a single polymer main chain plays the critical role in achieving TSCT emission. Solution-processed nondoped OLEDs based on P1 containing 5 mol% triazine acceptor exhibit blue electroluminescence with Commission Internationale de l’Eclairage (CIE) coordinates of (0.176, 0.269), and EQE of 12.1% as well as small efficiency roll-off of 4.9% at luminance of 1000 cd m$^{-2}$. Although the nonconjugated structure of polystyrene backbone is favorable for achieving blue emission, it is challenging to realize long wavelength emissions (green, yellow, and red) because of the interrupted electron delocalization. To address this issue, four kinds of polystyrene-based TSCT polymers (P3-P6, Figure 3) containing acridan donor and triazine acceptors with different electron-withdrawing ability are designed to tune the emission color.[62] For deep blue emission, acceptor having cyclohexane ring on triazine segment (P3) is used instead of triazine acceptor to reduce the electron-withdrawing ability. For green and red emission, triazine acceptors containing strong electron-withdrawing substituents of trifluoromethyl (P4), cyano (P5), and 4-cyanopyridyl (P6) are selected to increase the electron-withdrawing ability. It is found that by changing the electron-withdrawing ability of the acceptors, the CT emission band
can be shifted to blue or red region. For example, by using cyclohexane substituent to decrease electron-withdrawing ability of acceptor, the emission band is blue-shifted from 486 (P1) to 469 nm (P3). In contrast, the emission can be red-shifted from 542 to 577 nm and 628 nm from P4 to P5 and P6 by using trifluoromethyl, cyano, and 4-cyanopyridyl groups to increase electron-withdrawing ability. Moreover, the polymers exhibited promising PLQY up to 74% in solid state. It is noted that TSCT polymers can exhibit aggregation-induced emission (AIE) effect due to dipole-dipole interaction between donor and acceptor upon aggregation. The emission intensity for P4 is increased by 117 times from solution to aggregation states. Solution-processed OLEDs for P3, P4, and P6 in nondoped state exhibit blue, green, and red emission with peaks located at 455, 525, and 616 nm, respectively. To realize white emission, the acridan donor and two triazine acceptors are simultaneously introduced to one copolymer (P7), therefore two donor/acceptor pairs are formed to generate both blue (472 nm) and yellow (550 nm) TSCT emission from a single polymer. By fixing the acceptor contents at 5 mol% (triazine segment) and 0.4 mol% (cyano-substituted triazine segment), respectively, balanced blue and yellow emissions are achieved for white emission. Solution-processed OLEDs based on neat film of P7 exhibit EQE of 14.1% with CIE coordinates of (0.31, 0.42), which is efficient two-color white light for single polymers.

Triarylboranes are attractive building blocks for D-A emitters with CT emission because of their promising electron-accepting properties induced by vacant p-orbitals of boron atom that can participate in conjugation with aryl groups. Moreover, the electron-withdrawing ability of triarylboranes can be tuned by introducing substituents and bridging atoms, making them attractive acceptors for developing TSCT polymers. By using triphenylboranes with different substituents including hydrogen (H), fluorine (F), and trifluoromethyl (CF3) as acceptor, three kinds of TSCT polymers (P8, P9, and P10, Figure 4) are developed. These substituents are chosen because the electron affinity is increased in order of H, F, and CF3, leading to enhanced CT strength between donor and acceptor. By increasing electron-withdrawing ability of triarylborane acceptors, TSCT emission of the resulting polymers are red-shifted from 429 nm (P8) to 443 nm (P9) and

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Figure 3: TSCT polymers with full-color and white emission. EL spectra for P3-P6 (a), aggregation-induced emission (AIE) effect for P4 in THF/water solution (b), image of P3-P6 (c), and EL spectra for P7. Reprinted with permission from Ref. 62 Copyright 2019 Wiley-VCH.
483 nm (P10) in film state, accompanied by improved PLQY from 26% to 53%. The resulting polymers reveal AIE effect with the emission intensity increased by ∼33-folds from solution to aggregation state. Meanwhile, TSCT polymers containing acridan as donor and oxygen-bridged triphenylboranes as acceptors in the side chain of polystyrene backbone were prepared (P11-P13).[64] Compared to nonbridged acceptors, oxygen-bridged triphenylboranes are expected to enhance spatial interaction between donor and acceptor owing to their rigid and planar structures, leading to suppressed nonradiative decay process and improved PLQY. The resulting polymers containing tert-butyl, hydrogen, and fluorne groups show deep blue (444 nm) to sky blue (480 nm) emission, accompanied with PLQYs of 32-70% in film. In addition, AIE behaviors are also observed for these TSCT polymers with emission intensity enhanced by ∼27-folds for P13. Solution-processed nondoped OLEDs based on P13 exhibited blue emission with CIE of (0.16, 0.27), and promising efficiency with EQE of 15.0%.

Besides polystyrene-based ones, another kind of TSCT polymers are constructed with polynorbornene as nonconjugated backbone.[65] Unlike polystyrene-based TSCT polymers where donor and acceptor are attached to different repeating units and could motion freely relative to each other, polynorbornene-based TSCT polymers have two advantages. First, norbornene monomer itself can act as a rigid skeleton to fix donor and acceptor owing to their rigid and planar structures, leading to suppressed nonradiative decay process and improved PLQY. The resulting polymers containing tert-butyl, hydrogen, and fluorne groups show deep blue (444 nm) to sky blue (480 nm) emission, accompanied with PLQYs of 32-70% in film. In addition, AIE behaviors are also observed for these TSCT polymers with emission intensity enhanced by ∼27-folds for P13. Solution-processed nondoped OLEDs based on P13 exhibited blue emission with CIE of (0.16, 0.27), and promising efficiency with EQE of 15.0%.
face-to-face alignment) can be formed. For P14 containing acridan as donor, the long axes of acridan segment is perpendicular to adjacent triazine segment, leading to edge-to-face alignment between donor and acceptor. But in the case of P15 containing teracridan with second-generation dendron as donor, acridan segment is parallel to neighboring triazine acceptor, resulting in face-to-face alignment with interplanar distance of 3.52 Å. Compared to edge-to-face alignment with small oscillator strength ($f = 5.9 \times 10^{-5}$ (P14), the face-to-face alignment exhibits much larger oscillator strength ($f = 3.9 \times 10^{-3}$ for P15), implying that higher radiative transition rate can be achieved by face-to-face alignment, which is consistent with reduced density gradient analysis where the face-to-face alignment shows larger cofacial $\pi-\pi$ interaction region compared to edge-to-face alignment. Consequently, P15 with face-to-face alignment exhibits higher PLQY of 74% and RISC rate ($5.9 \times 10^6$ s$^{-1}$) than P14 with edge-to-face alignment (53% and $4.1 \times 10^6$ s$^{-1}$, respectively). Solution-processed nondoped OLEDs using TSCT polynorbornenes with face-to-face alignment exhibited blue emission with EQE of 18.8%, superior to those using the edge-to-face counterpart (EQE = 12.9%).

Moreover, TSCT polymers can also be utilized as host materials for white electroluminescence. First, TSCT polymers are composed of electron-rich donor and electron-deficient acceptor, making them capable to transport both holes and electrons in emissive layer. Second, the small $\Delta E_{ST}$ for TSCT polymers can maintain high triplet energy level
(T₁) and low singlet energy level (S₁), which is favorable for confining triplet excitons on emissive species and meanwhile keeping smooth carrier injection ability from adjacent layers. Third, TADF effect for the polymer as host enables up-conversion of triplet excitons to singlet ones, which can promote long-range (∼10 nm[75]) Förster resonant energy transfer (FRET) from host to dopant, resulting in lower triplet exciton density and reduced efficiency roll-off at high luminance.[76–78] By attaching phenylthienopyridine-based yellow phosphor (Ir(tptpy)2(acac)) into side chain of a blue-emitting TSCT polymer host containing acridan donor and triazine acceptor, single white-emitting polymer P16 is developed (Figure 6). By tuning content of phosphor to achieve partial energy transfer from the host to the phosphor, both blue delayed fluorescence from TSCT polymer host and yellow phosphorescence from Ir(tptpy)2(acac) are generated for warm white emission. The single-white-emitting polymer P16 containing 0.6 mol% phosphor exhibited CIE coordinates of (0.38, 0.43), together with low turn-on voltage of 3.0 V and maximum luminous efficiency (LE) of 51.9 cd A⁻¹ for solution-processed nondoped OLEDs. Remarkably, the polymers exhibit very small efficiency roll-off at high luminance. At a luminance of 1000 cd m⁻², the LE is kept at 50.9 cd A⁻¹, corresponding to efficiency roll-off of only 1.9%.

2.2 TSCT polymers with bottlebrush architecture

Bottlebrush polymers, consisting of linear backbone and macromolecular side chains, represent an important approach for creating the hierarchical nanomaterials. Typically, bottlebrush polymers have extended and wormlike conformations due to steric hindrance of the closely packed macromolecular side chains. This unique morphology and suppressed chain entanglement enabled the side chain spatially aligned around the polymer backbone, making them a promising platform for developing TSCT polymers.

In 2019, Hudson et al. reported the first bottlebrush polymers (P17-P19) with different sequence structures (random, miktoarm, and block architectures) by Cu(0) reversible-deactivation radical polymerization (Cu(0)-RDRP) (Figure 7) for achieving TSCT emission and TADF effect.[67] The random polymer (P17) has one kind of macromolecular side chain in which the donors and acceptors are copolymerized and randomly distributed, while the miktoarm polymer (P18) contains two kinds of macromolecular side chains consisting of either donors or acceptors. However, the block copolymer (P19) is composed of two blocks containing either donor-based macromolecular side chains or acceptor-based ones in each block sequence. This molecular design enables the investigation of effect of sequence distribution on TSCT emission for the resulting copolymers. It is found that PL spectra for random polymer (P17) and miktoarm polymer (P18) are dominated by CT emission at blue-green region (λmax = 490 nm) in solid-state film and aggregation state. In contrast, the diblock copolymer P19 exhibits almost no CT emission with the PL spectra appearing as the superposition of acridan and triazine segments. Similarly, fluorescence lifetimes of the bottlebrush copolymers are strongly dependent on sequential distributions. The random and miktoarm copolymers (P17 and P18) are dominated by delayed fluorescence in microsecond range (75.5–129 µs), indicative of TADF effect. In the case of block copolymer P19 with separated donor and acceptor regions, the long-lived delayed fluorescence is completely absent. These results demonstrate that the bottlebrush architecture can be a promising method for either enhancing or decreasing through-space donor-acceptor interactions in luminescent polymers by controlling distributions and morphology of donor and acceptor domains.

2.3 TSCT polymers with dendritic architecture

Dendrimers are well-defined tree-like structures that incorporate central core, branched functional groups, and multiple surface groups. Owing to the unique structures, dendrimers can combine the advantages of precise chemical structure and absolute molecular weight like small molecules, and good solution processability and morphological stability like polymers. Moreover, their core-shell structures can prevent concentration quenching and lead to high PLQYs in solid state, making them suitable for solution-processed OLEDs.

In 2019, our group reported the first example for TSCT dendrimer (P20, Figure 8) having circularly arrayed donor and acceptor segments in periphery of hexaphenylbenzene core.[68] Unlike conventional luminescent dendrimers...
containing emissive chromophores either in core or in dendrons, TSCT dendrimers contains no specific emitters but only alternatively aligned electron-donating and electron-accepting dendrons for through-space interaction. Theoretical calculations show that HOMOs of TSCT dendrimer are mainly distributed on acridan donors, but LUMOs are localized on triazine acceptors. Owing to the physically separated HOMO and LUMO distributions, small $\Delta E_{ST}$ of 0.0002 eV is observed for the dendrimer, leading to delayed fluorescence with lifetime of 3.54 µs. PL spectra of the dendrimer show CT emissions with red-shifted peaks relative to control compounds containing only donors or acceptors. Moreover, thanks to the nonplanar and propeller-shaped structure of hexaphenylbenzene core, this TSCT dendrimer is able to reveal AIE property through restricted intramolecular motion,[72] with $\sim$17-folds enhancement for emission intensity in aggregation state. Solution-processed OLED based on doped film of P20 (10 wt%) in hexa(4-(9,9-dimethylacridin-10-yl)phenyl)-benzene (Ac6) host shows EQE of 14.2% at maximum and 10.4% at luminance of 1000 cd m$^{-2}$. 

FIGURE 7 Schematic diagram and chemical structure for TSCT polymers with bottlebrush architecture (a and b), together with TADF and AIE effect (c and d). Reprinted with permission from Ref. 67 Copyright 2019 American Chemical Society.
FIGURE 8  Molecular structures and frontier molecular orbital distributions for TSCT dendrimer (a and b), together with the TSCT emission (c) and AIE effect (d and e). Reprinted with permission from Ref. 68 Copyright 2019 Royal Society of Chemistry

Recently, Yang et al. reported multi-(donor/acceptor) emitter with through-space/through-bond charge transfer by introducing three di-tert-butylcarbazole as donor and three difluorocyanobenzene as acceptor into phenyl core (P21, Figure 9).[69] This molecular design not only use the alternating arrangement of donor and acceptor at ortho-positions of phenyl ring to combine through-space and through-bond charge transfer emissions together, but also utilize multi-(donor/acceptor) architecture to promote efficient multichannel RISC process. The coexistence of through-space and through-bond charge transfer is supported by density functional theory calculation results that HOMO and LUMO are overlapped on both phenyl bridge and the interspace between donor and acceptor. Moreover, owing to the closely aligned high-lying singlet and triplet excited states, there are multichannel RISC processes for converting triplet excitons into singlet ones. The RISC rate constants are $(5.07 \pm 0.65) \times 10^5$ s$^{-1}$ for P21, much higher than the control emitters having fewer donor/acceptor units (P22 and P23, 0.12-2.27 \times 10^5 s$^{-1}$). It is found that P21 exhibits promising PLQY of 76% in neat film, giving the maximum EQE of 21.0% for solution-processed nondoped OLEDs with sky-blue emission.

3  |  CONCLUSION AND OUTLOOK

Since our group reported the first example for TSCT polymers with nonconjugated polystyrene backbone and spatial acridan donor/triazine acceptor architectures for achieving TADF effect in 2017, there have been some reports regarding the design and synthesis of new TSCT polymers and dendrimers, which have various nonconjugated polymer backbone and donor/acceptor architecture for solution-processed OLEDs. Their emission colors are able to cover the whole visible range from deep blue to red emission by regulating through space charge transfer strength, and the external quantum efficiencies of the OLED devices have reached 18.8% for linear polymers and 21.0% for dendritic ones, indicating they are promising for the development of solution-processed OLEDs.

Despite of the above progress, the development of TSCT polymers still faces several challenges. First, PLQYs of TSCT polymers are moderate (<80%) yet due to the lack of control for interactions between donor and acceptor. To address this issue, polymer backbone with fixed anchoring sites for confining donor and acceptor segments in not only close distance, but also controlled orientation (especially the cofacial alignment for donor and acceptor), should be developed to enhance the through-space $\pi-\pi$ interaction. Moreover, to suppress nonradiative decay losses caused by molecular motions, attentions should also be paid to the design of rigid and fused donor and acceptor segments, which can lock their conformations and restrict the free rotation and vibration. Second, TSCT polymers often suffer from broad emission band with full-width at half-maximum (FWHM) larger than 70 nm due to the structure relaxation of CT excited state. To improve color purity, sensitized fluorescent OLED devices by using TSCT polymers as sensitizer or assistant dopant for conventional fluorescent emitter could be explored to achieve narrowband emission. Third, as a kind of shortcomings for many luminescent polymers, TSCT polymers suffer from broad polydispersity for molecular weights and
structural uncertainty such as random sequence distribution and end-group effect. Under this circumstances, living polymerization methods with controlled and narrow-distributed molecular weights, regulable sequence structures, and tailorable end groups are highly desired for synthesis of TSCT polymers.\cite{67}

We believe that under the efforts of researchers in chemistry, physics, and material science, TSCT polymers will become one of the most important subjects in the future research field of luminescent polymers and solution-processed OLEDs. In addition, as a kind of $\pi$-stacked donor-acceptor system with long-lived delayed fluorescence (0.1-100 µs) derived from triplet states, TSCT polymers should also be useful in other applications such as oxygen sensing and time-resolved fluorescence imaging.

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CONFLICT OF INTEREST
The authors declare that there is no conflict of interest.

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