Aggregation-induced emission: Red and near-infrared organic light-emitting diodes

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Abstract
Red and near-infrared (NIR) organic light-emitting diodes (OLED) have gained remarkable interest due to their numerous applications. However, the construction of highly emissive emitters is hampered by the energy-gap law and aggregation-caused quenching (ACQ) effect. Whereas, aggregation-induced emission (AIE) materials could avoid the undesirable ACQ effect and emit bright light in aggregated state, which is one class of the most promising materials to fabricate high-performance OLED with a high external quantum efficiency and low efficiency roll-off. This review summarizes recent advances in red and NIR OLED with AIE property, including the traditional fluorescence, thermally activated delayed fluorescence, and hybridized local and charge transfer compounds. Meanwhile, the emphasis attention is paid to the molecular design principles, as well as the molecular structure-photophysical characteristics. We also briefly further outlook the challenges and perspective of red and NIR AIE luminogens.

KEYWORDS
aggregation-induced emission, luminous efficiency, organic light-emitting diodes, red and near-infrared emission
INTRODUCTION

Organic light-emitting diodes (OLED) have shown promising prospects in display and illumination fields due to the unique advantages such as self-emitting displays, transparency, fast response time, light weight, flexibility, and low-cost fabrication.\(^1\)–\(^{11}\) Over the last few decades, great progress has been made in achieving highly efficient OLED devices, resulting in the commercialization of television, smartphone, and wearable devices.\(^{12}\)–\(^{16}\)

Wherein, red light is not only an indispensable part of three primary colors for full-color displays and white OLED but also widely applied in the field of optical telecommunication, information secured devices, night-vision displays, and bioimaging.\(^{17}\)–\(^{24}\) Although there are rapid advances in organic light-emitting materials with various colors through rational molecular design,\(^{25}\)–\(^{40}\) the development of red and near-infrared (NIR) materials is much slower due to the intrinsic narrow bandgap and aggregation-caused quenching (ACQ) effect.\(^{41}\)–\(^{43}\)

Conventional red chromophores generally originated from nearly planar macrocyclic aromatic compounds with extended $\pi$-conjugation structure, such as polycyclic aromatic hydrocarbon (Figure 1A),\(^{44,45}\) or compounds with strong electron-donate and -accept units, such as electron-donor substituted pyran derivatives.\(^{46}\)–\(^{48}\) However, these red emitters are usually highly emissive in solution but weakly emissive in solid state, due to the effective dipole–dipole interactions and intermolecular $\pi$-stacking between chromophores with planar conformation in the aggregated state.\(^{49,50}\) For example, Nile red\(^{51}\) (Figure 1A) is a classical commercial red dye with high fluorescence quantum yield (67%) in dilute 1,4-dioxane solution, but almost no emission when aggregated. As the emissive layer works in a solid film state, the ACQ effect greatly hinders the practical application of red emitters in OLED.\(^{52}\) To solve the problem, a doping strategy is employed to suppress the molecular aggregation, but there are still inherent challenges.\(^{43}\)

First, it requires careful control of dopant concentration, which brings a lot of difficulties to adapt for mass production. Second, the doped devices would inevitably encounter performance degradation over time due to phase separation. Third, the high cost of fabricating doped devices. By contrast, the non-doped devices would not be haunted by these problems, indicating extensive

**FIGURE 1** Schematic illustration of aggregation-caused quenching (ACQ) and aggregation-induced emission (AIE). (A) Left: Nile red dye is highly emissive in the single-molecule state but none emissive in the aggregated state, right: molecular structure of typical ACQ compounds. (B) Left: Tetraphenylethene (TPE) is none emissive in the single-molecule state but highly emissive in the aggregated state, right: molecular structure of typical AIE compounds.
application value and great exploitation foreground. Thus, it is highly desirable to develop non-doped device systems.

In 2001, Luo et al.\textsuperscript{53} coined the concept of aggregation-induced emission (AIE) which revealed that some fluorophores with twist conformation emit dimly in solution but become strongly emissive upon forming aggregates. Since then, a large amount of experimental and theoretical research works were carried out to extend the scopes of AIE and explore the intrinsic mechanisms.\textsuperscript{40,54–58} Currently, the widely accepted AIE mechanism is the restriction of intramolecular motions,\textsuperscript{40,59,60} including restriction of intramolecular rotation, and restriction of intramolecular vibration. For instance, tetraphenylethene (TPE) is a typical propeller-like AIEgen\textsuperscript{61–63} which consists of four phenyl rings through a C═C double bond (Figure 1B). In the solution state, the free rotation of phenyl rings and tautomORIZATION of the central ethenyl double bond cause the quenching of the excited state via nonradiative transition. When the molecules aggregate together, compact molecular packing and π–π stacking interaction are restrained by the distorted molecular conformation, which avoids emission annihilation.\textsuperscript{64} Thus, the emission efficiency of the TPE state is largely enhanced via suppressing molecular motion and reducing the nonradiative transition rate. That is to say, AIEgens can efficiently suppress exciton annihilation in an aggregated state, which favors high efficient electroluminescence (EL) performance in non-doped OLED devices. In addition, the ACQ luminophores can be converted into AIEgens through the decoration of AIE moiety,\textsuperscript{42,56,65} providing a convenient strategy for building highly efficient luminescent molecules. Some representative AIEgens are listed in Figure 1B, such as TPB, HPS, DPDBP, and so forth, all have very distorted molecular configurations.\textsuperscript{66} Nevertheless, the majority of AIEgens emit efficiently in a relatively short wavelength region (less than 630 nm), and the deep red ones are still rare.\textsuperscript{4,67}

As AIE materials afford an excellent solution to overcome the troublesome ACQ effect, it has been one class of the most promising material to fabricate high-performance OLED with a high external quantum efficiency (EQE) and low-efficiency roll-off. So far, red and NIR AIEgens have been the best candidates for constructing efficient long-wavelength non-doped OLED. In this review, we aim to symmetrically summarize the recent advances of red and NIR AIEgens in the field of OLED. According to the luminous characteristics, the luminophores are divided into traditional fluorescence, thermally activated delayed fluorescence (TADF), and hybridized local and charge transfer (HLCT). Their basic luminescent mechanism, molecular design strategy, representative compounds, and the corresponding photophysical properties and device performance are discussed, respectively. Lastly, we further outlook the challenges and prospects of long-wavelength OLED.

2  |  FUNDAMENTAL PRINCIPLE AND WORKING MECHANISM

2.1  |  Luminous mechanism

In the EL process, light is produced by the recombination of holes and electrons in the light-emitting layer of OLED devices.\textsuperscript{68,69} According to spin-statistics, charge recombination is expected to produce singlet and triplet excitons in a ratio of 1:3.\textsuperscript{70} EQE is the most important parameter to evaluate the device efficiency, which can be calculated as follows:

,$\eta_{\text{EQE}} = \eta_{\text{out}} \times \Phi_{\text{F}} \times \eta_{\text{r}} \times \gamma = \eta_{\text{out}} \times \eta_{\text{IQE}},$

where $\eta_{\text{out}}$ is the light outcoupling factor (usually assumed to 0.22), $\Phi_{\text{F}}$ presents the photoluminescence (PL) quantum yield, $\eta_{\text{r}}$ is production efficiency of radiative transition excitons, and $\gamma$ is carrier balance factor.\textsuperscript{71} The transition of singlet exciton to the ground state yields the prompt fluorescence (Figure 2A), and the intersystem crossing (ISC) transition between states with different electronic multiplicity is spin-forbidden, resulting in upper limit internal quantum efficiencies (IQE) of 25\% theoretically. To obtain efficient fluorescent materials, the nonradiative transition of the singlet excitons should be suppressed, that is, the rate constant of fluorescence $(K_F)$ should be significantly higher than that of the nonradiative transition $(K_{\text{nr}})$. However, even if both $\gamma$ and $\Phi_{\text{F}}$ are close to 100\%, the maximum EQE of fluorescent emitters can only be up to 5.5\%, because the upper limit exciton utilization efficiency is 25\%.\textsuperscript{72}

Fortunately, thanks to the great effort of scientists, the efficiency upper limit was found to be able to break by directly utilizing triplet exciton or converting triplet exciton to singlet one.\textsuperscript{73–76} The directly radiative decay of triplet excitons results in phosphorescence, which requires phosphorescent emitter of organic metal complexes with the incorporation of transition metals, such as iridium (Ir), platinum (Pt), and ruthenium (Ru), which could largely promote the spin-orbital coupling (SOC) effect to enhance phosphorescence intensity. However, the large-scale commercialization of phosphorescent material is disturbed by the high price of these precious metals and the potential harm to the environment.\textsuperscript{77–81} The triplet exciton can also be utilized via converting to singlet exciton through the reverse intersystem crossing (RISC) process.
On the basis of the RISC channel, there are mainly three kinds of transition channels: TADF, HLCT, and triplet–triplet annihilation (TTA).

TADF is one of the most promising methods for harvesting triplet exciton, which involves the RISC process of exciton from T1 to S1 state with the aid of thermal energy from the ambient condition, then radiates as fluorescence (Figure 2B). Both singlet and triplet excitons can be utilized for light emission, resulting in 100% IQE of TADF emitters theoretically, which is comparable to that of phosphorescent OLED. In 2011, Endo et al. first reported the nearly 100% IQE in an OLED by using a purely organic TADF emitter and the high EQE of 5.3% almost broke up the theoretical limit of conventional fluorescent materials.

According to Hund’s rule, the S1 energy is considerably higher than that of T1, thus, the rate constant of ISC (KRISC) in conventional fluorescent materials is usually two to three orders of magnitude larger than that of RISC (KRISC), which forbids the RISC process of triplet exciton. The key point to realize TADF is to accelerate the RISC process. Furthermore, the fast RISC rate can not only increase exciton utilization, but also reduce the concentration of triplet excitons, which contributes to avoid device efficiency roll-off caused by TTA, singlet–triplet annihilation (STA), and triplet–polaron annihilation (TPA). From Boltzmann distribution:

\[ K_{RISC} = \exp \left( -\frac{\Delta E_{ST}}{k_BT} \right) \]

where \( \Delta E_{ST} \) is the energy gap between \( S_1 \) and \( T_1 \) state, \( k_B \) is Boltzmann’s constant, and \( T \) is the temperature. It can be found that \( K_{RISC} \) is in an inverse proportional to \( \Delta E_{ST} \), indicating that the \( K_{RISC} \) can be accelerated through reducing \( \Delta E_{ST} \). When the \( \Delta E_{ST} \) is sufficiently small (less than 0.1 eV), the energy barrier can be overcome by the aid of the environmental temperature. Moreover, \( \Delta E_{ST} \) is correlated to the electron exchange energy \( (J) \), which can be expressed as:

\[ \Delta E_{ST} = E_S - E_T = 2J \]

\( J \) depends on the electron density overlap between the highest occupied molecular orbital (HOMO) and the lowest unoccupied molecular orbital (LUMO):

\[ J = \int \int \phi_{HOMO}(r_1)\phi_{LUMO}(r_2) \frac{1}{|r_2 - r_1|} \phi_{HOMO}(r_2) d\mathbf{r}_1 d\mathbf{r}_2, \]

where \( \phi_{HOMO} \) and \( \phi_{LUMO} \) represent the spatial distributions of the HOMO and the LUMO, respectively, and \( r_1 \) and \( r_2 \) are the position vectors. Thus, the small \( \Delta E_{ST} \) can be realized through separating frontier molecular orbitals (FMOs). In molecular design, electron donor (D) and acceptor (A) are generally introduced simultaneously, in which D provides HOMO and A affords LUMO. The spatial separated D and A through molecular structure would give rise to the separated distribution of HOMO and LUMO, leading to small \( \Delta E_{ST} \).

Another way to achieve fast \( K_{RISC} \) is to separate HOMO and LUMO by multiple resonance effects. Different from the D–A-separated molecular configuration, the
multiple-resonance-induced TADF emitter is generated by combining and arranging atoms with opposite resonance effects in the same π-conjugated plan, featuring an alternating HOMO and LUMO distribution on electron withdraw and donate atoms respectively. Notably, benefiting from an appropriate overlap integral of FMOs, the small $\Delta E_{\text{ST}}$ and large oscillating strength ($f$) could be simultaneously achieved.\textsuperscript{96,97} Meanwhile, the unique mechanism can minimize the bonding and antibonding character, largely reducing molecular vibrionic coupling and vibrational relaxation via nonbonding molecular orbitals. As the result, these emitters have high color purity and photoluminescence (PL) efficiency photoluminescence quantum yield (PLQY) because of the extremely sharp emission peak with narrow full width at half maximum, exhibiting the enormous potential for OLED.

HLCT is a kind of hybrid or mixing state of locally excited (LE) and charge transfer (CT) states that the low-lying singlet- and triplet-excited states show LE features with a large energy difference, whereas the high-lying excited states have CT character with small energy splitting. Similar to TADF material, HLCT material can also harvest the triplet excitons and break through the spin-statistics limitation, achieving 100% IQE. However, the RISC process of HLCT occurs from high-lying triplet to singlet excited states ($T_n \rightarrow S_m, \ n \geq 2, \ m \geq 1$) (Figure 2C), which is clearly different from that of TADF (from $T_1$ to $S_1$).\textsuperscript{98} In 2012, Li et al.\textsuperscript{74} defined the RISC in a high-energy excited state as the hot exciton process. According to Kasha’s rule, the internal conversion (IC) process from $S_n$ to $S_1$ or $T_n$ to $T_1$ is much faster than the other processes.\textsuperscript{99} Thus, the high-order reverse intersystem crossing (hRISC) of hot exciton materials should be highly efficient when competing with the IC of $T_n \rightarrow T_1$. Considering that both RISC and IC rates are inversely proportional to the energy gap of two transition states, hot exciton materials should have a large $T_n$-$T_1$ energy gap to restrain IC and a relatively small $T_n$-$S_m$ energy splitting to accelerate hRISC. Besides, the efficient SOC of high-lying excited state can also increase upconversion efficiency,\textsuperscript{100} which contributes to reducing the accumulation of triplet exciton and avoiding the exciton annihilation caused by the long lifetime. Therefore, the HLCT OLED usually exhibits low-efficiency roll-off.\textsuperscript{101}

As for the TTA mechanism, under the theoretical framework, two triplet excitons in the lower energy can be converted into a singlet exciton with higher energy when the $\Delta E_{\text{ST}}$ ($S_1$-$T_1$) is large enough, generally requires $2T_1 > S_1$ (Figure 2D). The fluorescence generated by the TTA process is also called P-type delayed fluorescence, with the maximum IQE of 62.5% theoretically.\textsuperscript{102,103} However, due to the relatively high energy of the $S_1$ state, TTA is more common in deep-blue light OLED devices, which will not be discussed in this study. Besides, the technique of spin-polarized organic light-emitting diode (spin-OLED)\textsuperscript{104} even utilizes ferromagnetic electrodes as a bipolar organic spin valve to improve the EL efficiency.

### 2.2 Red and NIR emitters

Compared to the blue and green emitters, the red and NIR emitters require much smaller transition energy for light emission. Thus, reducing the energy ($E_g$) of $S_1$ is an essential condition to build red and NIR emitters, which is usually realized through extending electronic delocalization by increasing conjugation degree, leading to increased HOMO energy and reduced LUMO energy. The same effect can also be achieved by introducing suitable electron donors and acceptors to increase intramolecular charge transfer. Nevertheless, according to the energy gap law, the nonradiative transition rate ($K_{\text{nr}}$) increases exponentially as the $E_g$ reduces. The simplified mathematical expression for $K_{\text{nr}}$ could be described as follows:

$$k_{\text{nr}} \propto \exp\left(-\frac{\gamma \Delta E}{h\omega_M}\right),$$

where $\gamma$ is a term that in terms of molecular parameters, $\Delta E$ is the internal energy gap between the upper and lower states, $\omega_M$ is the frequency of the deactivating mode.\textsuperscript{105} Therefore, the low transition energy of red and NIR emitters would greatly increase $K_{\text{nr}}$ and results in low luminous efficiency, which has greatly obsessed the development of long-wavelength emitters with small $\Delta E$ for a long time. Besides, the red and NIR emitters would generally adopt the rigid molecular configuration to restrain $K_{\text{nr}}$ as much as possible.

### 2.3 Doped-device-based red and NIR emitters

Fortunately, significant progress has been made in red and NIR emitters currently for both PLQY and EL performance. Although lots of compounds did not exhibit the AIE property, they still achieve very high EQE through doping strategy, with the emission mechanism includes fluorescence, TADF, and HLCT. To provide a better presentation of structure difference between non-AIE and AIE emitters, some non-AIE compounds (or not study AIE property) are listed in Figure 3. The first doped red OLED was reported by Tang et al.\textsuperscript{106} in 1989, which was fabricated by fluorescence compounds of DCM1 and DCM2 with a maximum EQE of merely 2.3%. In recent
years, red and NIR OLED have achieved tremendous growth through elaborately molecular design, and high EQE mainly derives from TADF molecules. In 2014, Zhang et al. reported a D–A–D type red TADF molecule with a high $\Phi_F$ of 0.55 (614 nm) in a doped film. The $\Delta E_{ST}$ was largely reduced by increasing the distance between D and A through embedding phenylene linker, leading to a high EQE of 8.1% at a luminance of 100 cd/m$^2$ with EL at 624 nm. In 2017, Li et al. demonstrated a D–π–A type TADF emitter (TPA-QCN), which realized an EQE of 14.5% in a doped device with an EL peak at 644 nm. They further proposed supramolecular structure regulation that nonradiative decay of excited states can be reduced by proper molecular aggregation mode and the incorporation of rigid or planar constituent segments. With this strategy, in 2018, Chen et al. reported a red TADF emitter (DPXZ-BPPZ) with superior EQE of 20.1 ± 0.2% at 612 nm in doped devices. In 2019, Zhang et al. reported a red TADF emitter based on an acceptor PZCN, achieving a high EQE of 27.4% at 628 nm in the doped device, and the non-doped device exhibited EQE of 5.3% with an NIR EL at 680 nm. Recently, by using a similar structure to TPA-PZCN but with stronger electron-withdrawing ability via two additional CN groups on the acceptor segment, TPA-PZTCN exhibits more intense NIR EL with $\text{EQE}_{\text{max}}$ of 13.4% at 734 nm, and a long device operational lifetime (LT$_{95}$) of 168 h. In 2020, Zhao et al. found that the rational molecular packing mode in the neat film can efficiently suppress the exciton quenching and improve the carrier transport. They designed a deep-red TADF emitter $\rho$TPA-DPPZ with T-shape molecular structure, and the EQE of the non-doped bilayer device was up to 12.3% at 652 nm. In 2021, Chen et al. developed a series of red TADF emitters with similar D–A backbone but different peripheral groups, oDTB-PZ-DPXZ achieved an EQE of 20.1% in vacuum-processed OLED and 18.5% in a solution-processed OLED in the red region. There are also reports about HLCT OLED. In 2019, Chen et al. developed two red HLCT emitters, TPABTPA and TPABCHO, which enabled non-doped red OLED with EQEs of 11.1% at 608 nm and 5.0% at 640 nm, respectively. Wherein, TPABTPA possesses the highest EL efficiency among red hot exciton OLED.

FIGURE 3 Timeline of some typical non-aggregation-induced emission (AIE) (or not studied) red and near-infrared (NIR) organic light-emitting diodes (OLED)

3 | RATIONAL MOLECULAR DESIGN OF RED AND NIR AIEgens

Compared to non-AIE compounds, the AIE compounds adopt a spatial distorted molecular structure to prevent compact molecular interactions in the aggregated state. However, from the perspective of OLED device fabrication, the distorted structure of AIE compounds may hamper the charge transport, resulting in low carrier mobility and the orbital energy level would also affect...
carrier recombination in emitting layer. Therefore, lots of AIE emitters still adopt the host material to improve the EL performance. In this section, some typical red and NIR emitters with AIE character are discussed based on emission mechanisms of fluorescence, TADF, and HLCT.

### 3.1 Fluorescent red and NIR OLED

Due to spin forbidden, only singlet exciton can be utilized in fluorescent type OLED, leading to limit EL performance with a maximum EQE of 5.5%, which is much inferior to that of TADF and HLCT type OLED. Therefore, the reports on fluorescent organic red luminophores are relatively rare. The design of red AIE fluorescence usually adopts chromophore with strong electron donor and acceptor to achieve red emission, and the incorporation of AIE group to ensure AIE property. Perylene diimide derivatives (PDIs) is a famous red emitter with a largely extended conjugation degree, which have been wildly applied in various fields, such as solar cells, organic field-effect transistor, self-assembly, and fluorescent probes, and so forth, because of unique optoelectronic, thermal, chemical, and photochemical properties.\(^\text{116-118}\) However, their practical applications were greatly limited by the distinct ACQ effect. By decorating the PDI core with three twisty aromatic rings, Li et al.\(^\text{119}\) reported six red luminogens (Figure 4A) that change from ACQ to AIE. The bulky volume aromatic substituents play a crucial role in regulating the intermolecular \(\pi-\pi\) stacking, whereas the alkyl chains could improve the solubility of PDIs to improve the film morphology for solution-process OLED. Among them, STPH, DTPH, STRPH, and DTRPH still show ACQ effect, STTPE and DTTPE are AIE-active (Figure 4B), which attributes to the attachment of twisty AIE group, 4-(2,2-diphenyl-vinyl)-phenyl. Surprisingly, the EL performance of AIEgens is inferior to the four ACQ luminogens, maybe because the carrier mobility is impaired by the highly twisted substituents in STTPE and DTTPE. With a 1 wt% doping concentration, STRPH exhibited the highest EQE of 4.93% at 622 nm, approaching the theoretical limit of 5.5%, which is one of the best results among solution-process red OLED.

2,1,3-benzothiadiazole (BT) is also a well-known building block for constructing organic optoelectronic materials because of the planar molecular configuration and strong electron-withdrawing property.\(^\text{120,121}\) In 2015, Qin et al.\(^\text{122}\) designed two red AIEgens (TPE-TPA-BTD [TTB] and TPE-NPA-BTD [TNB]; Figure 5A) via taking BT as a central core to connect two TPE-substituted arylamine.\(^\text{122}\) Both of them exhibited emission at 617 nm with a high PLQY of 49% for TTB and 63% for TTN in solid film. The two compounds can serve as both emitting layer and hole-transporting layer (HTL) in non-doped bilayer OLED device, achieving EQE of 3.9% at EL emission of 604 nm for TNB, which is comparable with the device using \(N,N'\)-di(1-naphthyl)-N,N'-diphenyl-(1,1'-biphenyl)-4,4'-diamine (NPB) as the HTL. Lee et al.\(^\text{123}\) further developed two AIEgens of BT-2ATPE and BT-2NATPE (Figure 5B) by connecting TPE substituted arylamine to the BT directly.\(^\text{123}\) Despite that conjugation length is

![Figure 4](image-url)
shortened, two luminophores show much red-shift emission than that of TTB and TNB owing to a more intense ICT effect. The EL of the non-doped devices based on BT-2ATPE and BT-2NATPE reached to 684 and 682 nm, respectively. However, their EQE\textsubscript{max} was much lower than that of TTB and TNB, with merely 1.73% and 1.43%.

In 2018, a series of efficient red emitters consisting of TPE, BT, and phenanthro[9,10-d]imidazole were developed. Different numbers of tert-butyl and triphenylamine groups were introduced to regulate intermolecular interactions. Among them, t-BPITBT-TPATPE (Figure 6A) was designed with a tert-butyl-phenyl group on an imidazole unit and two triphenylamine groups on a TPE unit, exhibiting distinct AIE character. With the water fraction \( f_w \) in tetrahydrofuran (THF)/water mixtures increasing, apparently enhanced PL intensity was observed for t-BPITBT-TPATPE (Figure 6B). Theoretical calculation revealed that the HOMO mainly distributes on TPE and diphenylamine group, whereas that of the others is delocalized on the entire conjugated backbone. Consequently, TPATPE can efficiently consume the excited state energy by dynamic rotational or vibrational motions in the solution state. Moreover, the twisted intramolecular charge transfer effects resulted in more red-shifted emissions. The non-doped OLED of t-BPITBT-TPATPE showed a maximum EQE of 2.09% at 638 nm (CIE = 0.645, 0.345).

### 3.2 TADF red and NIR OLED

To realize red and NIR emission, TADF molecules would adopt electron donor with shallow HOMO energy and electron acceptor with deep LUMO energy, the well-separated donor and acceptor to guarantee the small \( \Delta E_{ST} \). Therefore, the electron donor and acceptor would not connect together directly, but is spaced by bridging groups, large torsional dihedral angle, through spatial isolation et al. In 2015, Wang et al.\textsuperscript{41} reported the first NIR TADF molecule TPA-DCPP (Figure 7) with V-shaped D–π–A–π–D configuration. 2,3-dicyanopyrazino phenanthrene (DCPP) served as acceptor to provide rigid configuration and strong electron-withdrawing capability, whereas diphenylamine acted as the donor for the outstanding hole-transporting capability as well as the large steric hindrance to diminish the ACQ effect. TPA-DCPP shows effective HOMO–LUMO separation and partial orbital overlap, resulting in a small \( \Delta E_{ST} \) of 0.13 eV and a large \( K_E \) of \( 9.0 \times 10^7 \) s\textsuperscript{-1}. Thus, TPA-DCPP film possessed a remarkable \( \Phi_{PL} \) of 14% with an emission peak at 708 nm. The EQE\textsubscript{max} of the non-doped OLED device reached 2.1% with a CIE coordinate of (0.70, 0.29). In the doped device, the maximum EQE was nearly 10% with EL emission at 668 nm, which is superior to most of the deep-red and NIR phosphorescent OLED with similar EL spectra.
In 2019, two orange to red TADF luminophores of NAI-BiFA and NAI-PhBiFA (Figure 8A) were reported by Peng et al.\textsuperscript{125} Wherein, naphthalimide (NAI) serves as an acceptor for not only strong electron-withdraw ability, but the planar rigid and easily modified architecture. Meanwhile, to endow the AIE effect, 2,6-diphenyl-4-toluene and arylamine donor unit with huge steric-hindrance was introduced into the NAI core. As previously mentioned, the insertion of a phenyl linker between D and A would further separate HOMO and LUMO and improve PLQY.\textsuperscript{108} Thus, NAI-PhBiFA was designed for comparison. Both emitters exhibited obvious AIE properties, which was demonstrated by PL spectra of them in CH$_2$Cl$_2$ and hexane solution (Figure 8B). Due to the active intramolecular rotation of the aryl rings, NAI-BiFA and NAI-PhBiFA emit weakly in CH$_2$Cl$_2$, however, when the hexane content increases gradually, the fluorescence intensity increases as well. In doped 4,4'bis(N-carbazolyl)-1,1'biphenyl (CBP) film, the $\Delta E_{ST}$ of NAI-PhBiFA (0.12 eV) is smaller than that of NAI-BiFA (0.15 eV), offering an EQE of 4.81% and 7.59% for NAI-BiFA and NAI-PhBiFA with a doping concentration of 5 wt% (Figure 8C,D), respectively. Nevertheless, probably due to triplet–triplet and singlet–triplet annihilations in the condensed neat film, the non-doped devices fabricated by NAI-BiFA and NAI-PhBiFA films showed poor EQE of 1.53% and 1.39%, respectively. In spite of this, the efficiency roll-off of non-doped OLED was notably smaller than that of doped OLED, indicating better efficiency stability.

In 2020, Wang et al.\textsuperscript{126} developed two D–A–D type red TADF emitters, TAT-DBPZ and TAT-FDBPZ (Figure 9A), by combining the donor of triazatruxene (TAT) and the acceptor of dibenzo-[a,c]phenazine (DBPZ) or fluorine-substituted dibenzo-[a,c]phenazine (FDBPZ). Both donor of TAT and acceptor of DBPZ possess a large...
\(\pi\)-conjugated plane structure, which delivers the delocalization of HOMO and LUMO to increase transition dipole moment and PLQY. The decoration of fluorine atoms could further increase the electron-acceptability of DBPZ, leading to more red-shift emission and shorter delayed fluorescence lifetimes. Owing to the large steric hindrance, a highly distorted structure is formed between TAT and DBPZ segment, leading to small \(\Delta E_{ST}\) and efficient suppression of intermolecular interaction. Moreover, both emitters showed AIE features with weak emission in THF, and strong emission as the water fraction increased, ensuring the high PLQY in the solid state (Figure 9B). The solution-processed doped OLED of TAT-DBPZ reached an EQE_max of 15.4\% with EL at 604 nm, which is superior to most of the solution-processed red TADF devices with the emission over 600 nm. Whereas, TAT-FDBPZ showed smaller \(\Delta E_{ST}\) and larger \(K_{RISC}\), achieving EQE_max of 9.2\% with a red-shifted emission at 611 nm accompanied by good efficiency stability. Benefited to AIE property, the non-doped solution-processed red TADF OLED was constructed with EQEs of 5.6\% and 2.9\% for TAT-DBPZ- and TAT-FDBPZ, respectively.
In 2020, Yang et al.\textsuperscript{127} reported two D–A type aggregation-induced delayed fluorescence (AIDF) emitters, pipd-BZ-PXZ, pipd-BZ-PTZ (Figure 10A), where a fused N-heterocycle (imid-azo[1,2-a]pyridin-2-yl(phenyl), pipd) and diarylketone acceptor served as acceptor, and phenoxazine (PXZ), phenothiazine (PTZ) group functioned as donors respectively. Owing to the embedded two nitrogen atoms, the pipd exhibited very strong electron-withdraw ability, which favors the ICT effect and red-shifted emission. In addition, the rigid planar conformation of pipd unit could increase PLQY by reducing the nonradiative energy dissipation in solid state. The rotatable phenyl group on the pipd favors the formation of twisted conformation, which is essential for realizing AIE and TADF properties simultaneously. As shown in Figure 10B,C, pipd-BZ-PXZ displayed intensified PL upon adding a large amount of water into THF solution, accompanied by a significant blue-shifted emission due to the ICT effect. Furthermore, pipd-BZ-PXZ and pipd-BZ-PTZ showed orange to red emission in solution and neat film, and moderate EL performance with the emission around 570 nm and EQE of 7.04% and 6.90% in nondoped devices, respectively. Thanks to the merits of the AIE property, the OLED device showed impressive efficiency stability. For instance, pipd-BZ-PXZ bears a small current efficiency roll-off of 2.3% at a brightness of 1000 cd/m², which is one of the best results among the orange-red OLED.

In the next year, they further introduced a CN-group into pipd-BZ-PXZ at different substituting sites to generate BenCN-PXZ and BDCN-PXZ (Figure 10D).\textsuperscript{128} The introduction of the CN group can not only facilitate the spin-flip of the ISC process but also suppress the nonradiative deactivation to enhance PLQY. Furthermore, owing to the stronger electron-withdraw nature of the acceptor group, the intramolecular CT effect was further enhanced, resulting in the red-shifted emission to a longer wavelength region. Compared to pipd-BZ-PXZ, the emission of the two new emitters red-shifted more than 30 nm. Meanwhile, their delayed lifetime (within 0.6 µs) is shorter than that of pipd-BZ-PXZ film of 0.8 µs, implying the promising application in OLED. The non-doped OLED with BenCN-PXZ and BDCN-PXZ as emitting layer showed orange-red to red emission at 592 and 610 nm with EQEmax of 4.94% and 3.71% (Figure 10E,F), respectively. Both of them showed a small EQE roll-off, especially for the latter one, with only 5.6% roll-off at 1000 cd/m². The doped OLED by employing 5 wt% BDCN-PXZ film as emissive layer also showed high current efficiency (CE), power efficiency (PE), and EQE up to 42.50 cd/A, 27.81 lm/W, and 13.75%. Notably, the device

![Figure 9](image-url)
fabricated by 20 wt% BDCN-PXZ demonstrated unprecedented small efficiency roll-off of 2.8% and 11.2% at the luminance of 1000 and 5000 cd/m², which is one of the best results among orange-red TADF OLED.

In 2018, Song et al. reported a series of binaphthalene-containing enantiomers with both AIE and TADF properties. The red emitter of R/S-BN-AF (Figure 11A) was designed by the decoration of 9,9-dimethyl-9,10-dihydroacridine (DMAC) unit. The single crystal illustrated the twisted conformations of S-BN-DCB with a dihedral angle of 74° between DMAC and phenyl ring (Figure 11B). The twisty configuration leads to weak π−π interaction in solid state, which contributes to the suppression of the exciton annihilation. Meanwhile, the abundant C−H⋯π and C−H⋯N interactions in the crystal lattice would rigidify the molecular conformation and lock the intramolecular rotations. However, the crooked conformation of the DMAC unit is harmful to TADF performance. Thus, S-BN-DCB possessed a low ΦPL of 5.3% in the neat film. Owing to the chiral binaphthol skeleton, both cotton effects and circularly polarized luminescence for R/S-BN-AF in toluene solution and films were detected. Multilayer CPOLED using 5 wt% S-BN-DCB: CBP doped film and the neat film as emitters exhibited EQE of 1.5% and 0.5%, with EL dissymmetry factor (gEL) of 0.016−0.027 and 0.084−0.091, respectively. Additionally, because of the stronger AIE property in the neat film, the non-doped CPOLED showed a much smaller efficiency roll-off and larger gEL than that of the doped one.
3.3 HLCT red and NIR OLED

HLCT compounds have a unique electronic structure that can utilize triplet exciton for light emission, that is, triplet exciton is converted to singlet one through the hRISC process. Therefore, the exciton radiative transition channel and the RISC channel are separated, which avoids the contradiction between the large overlap of FMOs for large oscillator strength and the small overlap for small energy splitting. Different from the TADF molecule, the S1 state of the HLCT molecule can be an LE state, contributing to improving PLQY. The hRISC process would also reduce the accumulation of T1 state exciton, effectively diminishing the exciton annihilation. Thus, the HLCT OLED usually has a relatively small efficiency roll-off. At present, the electron acceptor with the particular electronic structure for red HLCT compound is relatively rare, mainly includes benzothiadiazole (BZ) and naphthothiadiazole (NZ), whereas the AIE effect is derived from the distorted molecular configuration.

In 2014, Yao et al.\textsuperscript{131} reported a butterfly-shaped NIR D-A luminophore, PTZ-BZP (Figure 12A), where PTZ served as D and benzothiadiazole served as A. The density functional theory (DFT)-optimized geometry indicated dihedral angle between D and A (145°), and in twisty PTC group (142°), leading to AIE property of PTZ-BZP. Additionally, the relatively planar conformation might cause considerable orbital overlap and further enhance the K\textsubscript{T1} and PLQY. The PTZ-BZP film displayed a broad NIR PL emission around 700 nm with a quantum efficiency of 16%. The non-doped NIR OLED displayed EQEmax of 1.54% and brightness of 780 cd/m\textsuperscript{2}. Meanwhile, the OLED exhibited a low-efficiency roll-off, in which the EQE was as high as 1.17% even at a high current density of 300 mA/cm\textsuperscript{2}.

As shown in Figure 12B, the energy level of S1 state was almost identical to that of T3 state (\(\Delta E_{ST} \approx 0\) eV), whereas the energy gap between T3 and T2 state is relatively large (0.76 eV), which ensured the much faster hRISC process than IC process. Thus, the efficient hRISC provided by the hot-exciton process (T3 \(\rightarrow\) S1) is responsible for the high radiative exciton ratio of 48% in the OLED device.

In 2017, Liu et al.\textsuperscript{132} developed a red HLCT emitter NZ2AC (Figure 12C) by incorporating NZ and DMAC unit, and phenyl \(\pi\)-bridge into a D-\(\pi\)-A-\(\pi\)-D molecular architecture. NZ group possesses an inherent electronic structure in which there is a large energy difference between the triplet-excited state platforms (such as T2 \(\rightarrow\) T1), making it an ideal unit for constructing HLCT materials. From Figure 12D, the nearly
vertical dihedral angle between the DMAC and the phenyl bridge, and a wide angle between the NZ unit and the phenyl bridge demonstrate the twisty configuration of NZ2AC, leading to the separated HOMO and LUMO distribution on DMAC and NZ units, respectively. Moreover, the $\Delta E_{ST}$ is up to 1.27 eV and the $T_2$–$T_1$ energy gap is as high as 1.76 eV, resulting in an unfavorable RISC process from $T_1$ to $S_1$ and limited IC from $T_2$ to $T_1$. By contrast, there’s a small energy gap between $T_2$ and $S_2$ (0.17 eV), which accelerates the hRISC from $T_2$ to $S_2$. Meanwhile, NZ2AC possesses typical AIE feature, in which the PL intensity gradually increased as the water fraction rose (Figure 12E). The doped OLED (8 wt% in CBP) showed a red emission at 612 nm with EQE$_{\text{max}}$ of 6.2%, whereas the non-doped OLED displayed a deep-red emission at 663 nm with an EQE$_{\text{max}}$ of 2.8%. Moreover, due to the HLCT and AIE characters, both of the doped and non-doped OLED showed impressive low-efficiency roll-off, and the EQE remained high values of 3.0% and 2.3% at the luminance of 5000 cd/m$^2$, respectively.

In 2020, Tang et al. constructed three deep red luminogens (TNZPPI, TNZtPPI, and TNZ2tPPI) (Figure 13A) by 1,2-diphenyl-1H-phenanthro[9,10-d]-imidazole (PPI) unit and TPA-modified NZ (TNZ). All of them have both HLCT and AIE properties. PPI exhibits a large planar structure and bipolar characteristic, which helps to enhance the carriers’ injection or transport capability as well as the brightness of target molecules. To demonstrate the importance of the planar group of PPI unit, TNZtPPI and TNZ2tPPI were modified with tert-butyl group with different steric effect groups to inhibit intermolecular stacking between phenanthrene-based planes. As a consequence, TNZPPI realized the highest $\Phi_{\text{PL}}$ of 38%, maximum radiance of 36,027 mW/(Sr m$^2$), and EQE$_{\text{max}}$ of 2.48% with EL peak of 686 nm in non-doped OLED (Figure 13B,C), which was significantly better than tert-butyl substituted counterparts. The doped devices based on 10 wt% TNZPPI in CBP host also showed superior EL performance with EQE$_{\text{max}}$ of 6.83% (648 nm). The corresponding CIE of EL reached the requirement of standard saturated red light (SSRL) defined by the National Television System Committee (NTSC), which is the highest efficiency for SSRL devices at present.

In the same year, they reported another series of deep red HLCT AIEgens on the basis of the TNZ unit (Figure 13D). Three typical fused aromatic groups (pyrene [Py], phenanthrene [Phe], and naphthalene [Np]) with different $\pi$-conjugated extension were introduced to TNZ unit to produce PyTNZ, PheTNZ,

**FIGURE 13** (A) Chemical structure of TNZPPI, TNZtPPI, and TNZ2tPPI; (B) radiance of TNZPPI, TNZtPPI, and TNZ2tPPI versus voltage; (C) EQE versus brightness of TNZPPI, TNZtPPI, and TNZ2tPPI in OLED devices, the inset presents EL spectra of TNZPPI; (D) chemical structure of TNZ, PyTNZ, PheTNZ, NpTNZ; (E) calculated $T_1$ and $T_2$ energy levels of separated units and compounds based on the ground-state conformation; (F) PL spectra of NpTNZ in DMF/water mixtures with different water fractions. Reproduced (Adapted) with permission. Copyright 2020, Royal Society of Chemistry. EL, electroluminescence; DMF, N,N-dimethylformamide; EQE, external quantum efficiency; PPI, 1,2-diphenyl-1H-phenanthro[9,10-d]-imidazole; OLED, organic light-emitting diode; PL, prompt fluorescence; TNZ, TPA-modified NZ
NpTNZ, aiming to regulate singlet/triplet energy levels distribution. The $T_1$ energy of Py was lower than that of TNZ, leading to a smaller energy gap between $T_2$ and $T_1$ in PyTNZ. Whereas the $T_1$ energy of Np and Phe was higher than the $T_2$ of TNZ, resulting in a higher $T_2$ energy level and larger triplet energy gap between $T_2$ and $T_1$ for PheTNZ and NpTNZ (Figure 13E). Therefore, PheTNZ and NpTNZ exhibited a more efficient hRISC process and higher PLQY, with deep red emission around 642–678 nm in non-doped devices, which was close to the standard red emission evaluated by CIE coordinates. Meanwhile, the four emitters show typical AIE activity. For example, NpTNZ emits dimly in dilute $N$, $N$-dimethylformamide (Figure 13F), but exhibits a strong fluorescence signal when water fraction ($f_w$) increases. However, once $f_w$ reaches 60%, the emission intensity starts to decrease as the formation of molecular aggregate. The non-doped devices displayed high EQEs up to 2.63% and 2.20% for NpTNZ and PheTNZ, respectively. Additionally, the devices enjoyed stable efficiency roll-off owing to the “hot excitons” process and AIE property.

Recently, Zhang et al. further developed four BZ-based emitters of TBAN, TBT, TBAT, and TABAT (Figure 14A) by taking TPA as the donor, BZ as the acceptor, and anthracene (AN) as a bridge. The incorporation of TPA not only offered strong electron-donate ability but effectively reduced $\pi-\pi$ stacking and restricted molecular motions in the aggregated state, which favored for AIE effect. BZ is a rigid electron-withdraw group that has been widely used for constructing red emitters, but AN endows the target molecules with high fluorescent efficiency and good electron-transport ability. By manipulating the building units, these four compounds are designed with $D-A-\pi$, $D-A-D$, $D-A-\pi-D$, and $D-\pi-A-\pi-D$ structures with different excited-state properties.

![Chemical structure of TBAN, TBT, TBAT, and TABAT; (B) the PL spectra and photographs of TBAT in THF/water with different water fractions. Reproduced (Adapted) with permission. Copyright 2021, Royal Society of Chemistry. PL, photoluminescence; THF, tetrahydrofuran](image-url)
Probably caused by the similar molecular structure with NpTNZ, all the four compounds also exhibit changes in PL spectra depending on the water fraction. When \( f_w \) is smaller than 60%, the PL intensity decreases with the addition of water, probably attributed to the increased polarity of water/THF solution (Figure 14B). Whereas, as the formation of aggregated states at a high \( f_w \) from 60% to 99%, the PL intensity gradually enhances. The non-doped OLED showed orange to red emission from 580 to 608 nm, accompanied by high EQE and negligible efficiency roll-off. Among them, TBAN possessed the best EL performance, realizing a maximum EQE of 5.7% with EL at 596 nm. Moreover, attributed to the excellent hole/electron transport property, the four compounds could act as both hole-transport and emitting layers in one single device. The device based on TBAN exhibited high efficiency, which was comparable to that of the device with NPB as a hole-transport layer.

4 | SUMMARY AND OUTLOOK

Red and NIR light-emitting materials play an indispensable role in fabricating full-color and white light OLED. In the last couple of years, red and NIR AIEgens have attracted great attention due to the high PL efficiency in the aggregated state, which has become the most promising candidate for fabricating non-doped OLED devices. In this review, we discuss the basic knowledge of luminescent mechanisms and the progress of the AIEgens in the field of red and NIR OLED. The first generation of fluorescent OLED can only utilize singlet exciton for light emission. Although the highest EQE of red AIE fluorescent OLED has approached the theoretical limit of 5.5%, it is far from the satisfaction for commercialization. To get access to the remaining excitons in triplet states, OLED based on TADF and HLCT materials have achieved high EQE that is comparable to phosphorescent materials, and the device efficiency of TADF-OLED is generally superior to that of HLCT counterparts. Up to now, the maximum EQE of red AIE TADF-OLED has been advanced to 15.77%, whereas only 6.83% for HLCT-OLED. However, the HLCT device exhibits relatively low-efficiency roll-off because of the much lower concentration of \( T_1 \) excitons resulting from the fast hRISC process and the inhibited IC of \( T_n-T_1 \). Despite some achievements that have been made in highly efficient red and NIR AIE OLED, the low EQE is still the bottleneck. Furthermore, serious efficiency roll-off, broad light-emission spectrum, high fabrication cost, and short service lifetime are still challenging issues in the device application. To obstacle the dilemma, a more deliberate molecular design strategy and engineering means of red and NIR compounds with high EL performance should be carried out. For TADF materials, the reduced triplet exciton lifetime accompanied with enhanced RISC rate can effectively reduce the triplet exciton density to alleviate efficiency roll-off. Besides, TADF compounds with multiresonance effect exhibit advantages of narrow-emission spectrum, high luminous efficiency, and so forth, which deserves more attention to develop highly efficient luminophore. For HLCT compounds, the exploration of a more efficient acceptor unit should contribute to improving EL efficiency. Lastly, on the consideration of relatively shallow HOMO energy and deep LUMO energy of red and NIR luminophore, the hole/electron transport materials with matched energy levels should be investigated to further improve EL efficiency.

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CONFLICT OF INTERESTS

The authors declare no conflict of interest.

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