Allochroic thermally activated delayed fluorescence diodes through field-induced solvatochromic effect

Chunmiao Han,* Chunbo Duan,* Weibo Yang, Mingchen Xie, Hui Xu†

Allochroic organic light-emitting devices (AOLEDs) characterized by field-dependent emissive color variation are promising as visible signal response units for intelligent applications. Most of the AOLEDs were realized by changing their recombination zones or inter- and intramolecular energy transfer, rendering the limited repeatability, stability, and electroluminescence (EL) performance. We report a novel thermally activated delayed fluorescence (TADF) diode that featured a successive and irreversible emission color change from bluish green to deep blue during voltage increase, which uses the significant influence of host polarity on the emission color of TADF dyes, namely, solvatochromic effect. Its host 3,6-di-tert-butyl-1,8-bis(diphenylphosphoryl)-9H-carbazole (tBCzHDPO) was designed with remarkable field-dependent polarity reduction from 7.9 to 3.3 D by virtue of hydrogen bond–induced conformational isomerization. This TADF device achieves the best EL performance among AOLEDs, to date, with, for example, an external quantum efficiency beyond 15%, as well as the unique irreversible allochroic characteristic for visible data storage and information security.

INTRODUCTION
Allochroic organic light-emitting devices (AOLEDs) are one kind of unique OLED with variable emission color dependent on applied voltages; therefore, they can convert electrical signals to light color signals (1, 2). On that basis, AOLEDs are perfect for naked-eye information recognition, making them promising in nearly all display applications. The early AOLEDs were designed with two emissive layers (EMLs) with different colors, whose recombination zones can be switched or shifted between EMLs for the electroluminescent (EL) spectral variation (3–6). In contrast, through restraining energy transfer, Berggren et al. (7) achieved the ordinal emissions from different domains in EMLs of polymer blend–based OLEDs, forming another kind of AOLEDs with changeable intra- and/or intermolecular energy transfer. In actuality, it is a common phenomenon for OLEDs that recombination location shift (RLS) and energy transfer modulation (ETM) during voltage increase are often accompanied by slight spectral change due to its optical effect and emission ratio variation (8–10). However, these allochroic devices inevitably suffered from poor repeatability and weak EL performance. Therefore, in recent years, some new strategies were developed to realize field-induced allochroic behaviors, including emitters with multiple radiative transition channels [for example, carbon dots (11)] and voltage-dependent dynamic molecular structure rearrangement (12). Nevertheless, the involvement of excitons in these allochroic processes inevitably exacerbates quenching and reduces EL performance and repeatability. The great challenge is still that allochroic devices with applicable EL performance, such as brightness beyond 1000 cd m⁻² and external quantum efficiencies (EQEs) of more than 10%, are absent.

Thermally activated delayed fluorescence (TADF) devices rapidly gained attention owing to their advantages in 100% exciton harvesting, low cost, and environmental friendliness (13), in which emitters are characterized by donor-acceptor systems with strong intramolecular charge transfer (CT) for triplet-singlet exciton upconversion through reverse intersystem crossing (14–18). Within 10 years of the first reported TADF diode, the performance of TADF devices is already comparable to electrophosphorescence analogs, displaying their great potential for practical applications (19–22). It is noteworthy that the emissions from all TADF dyes show the remarkable bathochromic shifts along with the increase of the solvent polarity, namely, the so-called solvatochromic effect, which should be attributed to the stronger stabilization effect of higher polar solvents on their CT-featured excited states in contrast to ground states (Scheme 1A) (23). In OLEDs with doping-type EMLs, their host matrices actually serve as the solid solvents of emitters; therefore, the molecular polarities of host materials should markedly influence the emission colors of TADF devices (24). The remarkable emission bathochromic shifts of TADF diodes when slightly increasing doping concentrations, even from 1 to 3%, actually reveal the high sensitivity of TADF emission to environmental polarity (25). In contrast to their phosphorescent and fluorescent counterparts, TADF dyes are featured with efficiently radiative CT excited states, establishing the basis for their potential applications in high-efficiency AOLEDs. In our previous work, a significant bathochromic shift of emission peaks from 460 to 472 nm, corresponding to deep blue and greenish blue, were observed for conventional blue TADF dye bis[4-(9,9-dimethyl-9,10-dihydroacridine)-phenyl]sulfone (DMAC-DPS)–based devices (26) when using host materials with gradually increased polarities, which inspired us to develop TADF AOLEDs on the basis of solvatochromism (27). Herein, we report 3,6-di-tert-butyl-1,8-bis(diphenylphosphoryl)-9H-carbazole (tBCzHDPO) as a host with hydrogen bond–controlled polarity variation to fabricate a proof-of-concept TADF AOLED with the unique host-leading irreversible green-to-blue emission color change and the state-of-the-art EL performance (for example, the maximum EQE beyond 15%) (Fig. 1A), paving a way to practically applicable AOLEDs.

RESULTS

Polarity bistability

tBCzHDPO is designed as a carbazole–phosphine oxide (PO) hybrid. The intramolecular hydrogen bond (IHB) between N–H of its carbazole core and the oxygen atoms of its two diphenylphosphine oxide (DPPO) groups can fix its conformation as an endo type with stronger thermodynamic stability, which is confirmed by its single-crystal structure. Theoretically, tBCzHDPO can adopt three main conformation types with two outward, one outward–one inward, and two inward P=O...
bonds, corresponding to symmetrical and asymmetrical exo types and endo types, respectively. However, the symmetrical exo-type conformer is the most thermodynamically unstable because of its high total energy and large energy barrier of conformation transformation (fig. S1). In this case, the actual preferential conformations of tBCzHDPO are the endo and asymmetrical exo types. The latter is referred to simply as exo type. The density functional theory (DFT) simulation indicates the low polarity of endo-type tBCzHDPO by a level of ~3 D. In contrast, its exo-type conformation with a markedly increased polarity of ~8 D is the major conformer in as-deposited solid films on account of its two equivalent conformational isomers (Fig. 1B, fig. S1A, and table S1). The total energy of its endo conformer is 11.7 kJ mol$^{-1}$, lower than that of the exo conformer, revealing the higher thermodynamical stability of the former. Furthermore, the energy barrier of ~15 kJ mol$^{-1}$ for exo-to-endo transformation is surmountable. Therefore, tBCzHxPO and its mono-PO analog 3,6-di-tert-butyl-1-(diphenylphosphoryl)-9H-carbazole (tBCzHSPO), collectively named tBCzHxPO, are designed with IHB-controlled binary polar states. The main difference is that the polarities of the exo and endo conformers for tBCzHSPO are comparable as 3 to 4 D.

**Polar state transformation**

Time-dependent DFT (TDDFT) calculation of the nature transition orbitals (NTOs) (28) in singlet excitations for tBCzHSPO and tBCzHDPO indicated the influence of conformation variation on electronic characteristics of their exo- and endo-type isomers (fig. S2). For ground state ($S_0$) → singlet excited state ($S_1$) transitions, the comparison between the exo and endo conformers shows the decreased contributions from DPPO groups to their “particle” locations, giving rise to a remarkable $S_1$ energy reduction by 0.21 and 0.08 eV, corresponding to red shifts of 19 and 8 nm, respectively, which is in accord with DFT-simulated frontier molecular orbital (FMO) energy gaps (fig. S3). The solid-state emissions of tBCzHxPO vacuum-evaporated films before and after annealing show the identical situation. Photoluminescence (PL) spectra of the as-prepared films are peaked at 392 and 385 nm for tBCzHSPO and tBCzHDPO, respectively. After annealing at 200°C for 30 min, the remarkable emission bathochromic shift by ~10 nm can be observed for these films (fig. S4B). In this sense, the shorter emission peak wavelengths of the as-prepared tBCzHxPO films before annealing indicate the predominant ratio of their exo-type isomers with the higher $S_1$ excited energy. In actuality, when preparing thin films through high-temperature evaporation and rapid cooling, it is rational that the high-energy condition for evaporation is beneficial to overcome the rotational energy barrier. Therefore, the exo conformers should be dominant in the as-deposited films according to statistics. Significantly, the bathochromic emissions of annealed tBCzHxPO films manifest the exo-to-endo transformation by heating, ascribed to the higher thermodynamic stability of their endo-type isomers. The role as driving force for heat is further demonstrated with differential scanning calorimetry, by which tBCzHxPO films reveal the distinct glass-state transition accompanied by the exo-to-endo transformation (fig. S5 and table S1). To figure out the effect of IHB on conformation variation, the solid-state PL spectra of the vacuum-evaporated films based on the N-methyl–substituted derivative 3,6-di-tert-butyl-1-(diphenylphosphoryl)-9H-carbazole and 3,6-di-tert-butyl-1,8-bis(diphenylphosphoryl)-9-methyl-carbazole (tBCzMxPO) (29) without IHB were simultaneously measured to show the almost unchanged emissions before and after annealing, despite the similar exo-to-endo $S_1$ excited energy reduction and thermodynamic stabilization of tBCzMxPO (fig. S2). Therefore, IHB is the crucial force to fix endo-type conformations of tBCzHxPO during annealing and to induce monodirectional exo-to-endo transformation.
It can be expected that conformation switch of tBCzHDPO between its exo and endo isomers in the doped thin film would markedly change the matrix polarity of its dopants. To confirm the possibility of conformational transformation under an electrical field, we fabricated tBCzHxPO-based light-emitting devices with the configuration of indium tin oxide (ITO)/MoO3 (6 nm)/4,4′-bis[N-(1-naphthyl)-N-phenylamino]-1,1′-biphenyl (NPB) (40 nm)/1,3-bis(N-carbazolyl)benzene (mCP) (5 nm)/tBCzHxPO (20 nm)/bis{2-[di(phenyl)phosphino]phenyl}ether oxide (DPEPO) (5 nm)/4,6-bis(diphenylphosphoryl) dibenzothiophene (DBTDPO) (30 nm)/LiF (1 nm)/Al (100 nm) through vacuum evaporation, in which NPB and DBTDPO were adopted as hole-transporting layer and electron-transporting layer (ETL), respectively, whereas mCP and DPEPO with the appropriate FMO energy levels were used to improve the carrier injection and the confinement of charge carrier recombination in tBCzHxPO layers (scheme S2). Along with the voltage increase from 5.5 to 9.5 V, both tBCzHSPO and tBCzHDPO showed the voltage-dependent EL spectra with red shifts of 12 and 24 nm (Fig. 1C). As indicated by optical analysis, exo-type conformations of tBCzHxPO are dominant in their as-deposited films. The joule heat generated by electricity is basically proportional to the product of voltage and current density (J), namely, power density, therefore facilitating the exo-to-endo transformation as the same mechanism of the annealing effect on tBCzHxPO films.

Fig. 1. Molecular design of tBCzHxPO host with transformable binary polar states. (A) Single-crystal structures of tBCzHxPO, indicating the preferential endo structures with IHBs at thermodynamic stable states. (B) Conformation transformation between exo- and endo-type isomers of tBCzHxPO driven by hydrogen bond formation and the resulted polarity and energy variation. (C) EL spectra of tBCzHxPO-based OLEDs in a voltage range of 5.5 to 9.5 V with an interval of 0.5 V. (D) Photoluminescence (PL) spectra (inset) and time decay curves of emissions from vacuum-evaporated DMAC-DPS–doped tBCzHxPO and tBCzMxPO thin films [100-nm thickness, 10 weight % (wt %)].
To further verify the effect of electrical field on conformation variation of tBCzHxPO, we fabricated their single-layer devices with configuration of ITO/MoO$_3$ (6 nm)/tBCzHxPO (100 nm)/LiF (1 nm)/Al (fig. S6B). For the first circle of voltage increase from 0 to 20 V, the typical semiconductor volt-ampere characteristics were recognized with conductive voltages of 4.3 and 7.9 V for tBCzHSPO and tBCzHDPO, respectively. Then, in the second circle, their conductive voltages decreased to 2.8 and 1.5 V, respectively, and their $J$ markedly increased, reflecting improved ordered intermolecular packing after the first circle. Commonly, the hole and electron injecting abilities are roughly determined by the energy levels of the highest occupied and the lowest unoccupied molecular orbitals (LUMOs), respectively. In this sense, tBCzHDPO with the deeper LUMO should be superior to tBCzHSPO in electron injection (figs. S3 and S6A). However, the higher conductive voltage of tBCzHDPO-based devices in the first circle reflected the equally crucial influence of aggregation characteristics on carrier injection in the thick films. Besides the reduced FMO energy gaps of the endo-type conformers for carrier injection balance, in contrast to the exo-isomers, the endo types of tBCzHxPO are more symmetrical to enhance the regular alignment and intermolecular interaction for the improvement of carrier injection and transport, indicating the correlation between the increased $J$ in the second circle and the exo-to-endo transformation in the first circle.

**Photophysical properties**

tBCzHxPO show the optical properties in dilute solution almost identical to those of tBCzMxPO (fig. S4A) (29). Their high triplet excited energy ($T_1$) beyond 2.9 eV can support the positive energy transfer to DMAC-DPS, rendering the pure blue emissions and high photoluminescence quantum yields beyond 70% from their DMAC-DPS-doped vacuum-evaporated thin films (Fig. 1D). It is noteworthy that the PL emission peaks of DMAC-DPS in tBCzHDPO and 3,6-di-tert-butyl-1,8-bis(diphenylphosphoryl)-9-methyl-carbazole (tBCzMDPO) matrices are 8 nm longer than those in tBCzHSPO and 3,6-di-tert-butyl-1-(diphenylphosphoryl)-9-methyl-carbazole (tBCzMSPO). These bathochromic shifts of dopant emissions rationally manifest
the existence of the high-polarity states in tBCzHDPO and tBCzMDPO films and their significant influences on emission color. Furthermore, the emission profiles of these films are only dependent on the DPPO group numbers of their host matrices. In contrast to tBCzHxPO, without influence of IHB, the conformation population of tBCzMxPO should roughly follow the statistic probability. Therefore, the exo-type conformation of tBCzMDPO would be the major conformers in its films, whose polarities are almost the same as that of exo-type tBCzHDPO (fig. S7). In this sense, the identical emission of DMAC-DPS in tBCzHDPO validates the predominance of the exo-type isomers for the host in its as-deposited films, in accord with the situation of its nondoped films.

Device performance

Inspired by the electric field–induced conformation variation of tBCzHxPO, we created five-layer devices with the configuration of ITO/MoO3 (6 nm)/NPB (40 nm)/mCP (5 nm)/host:DMAC-DPS (10 wt %, 20 nm)/DPEPO (5 nm)/DBTDPO (30 nm)/LiF (1 nm)/Al (100 nm) in which mCP and DPEPO served as exciton-blocking layers (Fig. 2A and scheme S2). tBCzHDPO and tBCzMDPO endowed their devices with a reduced onset voltage of 3.0 V, 0.5 V lower than those of tBCzHSPO- and tBCzMSPO-based devices, which were in accord with the deeper LUMO of the former for more efficient electron injection (Fig. 2B and table S2). At a voltage range of 4.0 to 4.5 V, for commercial lithium batteries, the luminance of tBCzHDPO- and tBCzMDPO-based devices can increase to an applicable range of 25 to 100 cd m⁻². tBCzHDPO realized the highest efficiencies among these hosts, with maxima of 30.4 cd A⁻¹ for current efficiency, 38.2 lumen W⁻¹ for power efficiency, and 15.2% for EQE, which were comparable to the five-layer DMAC-DPS–based counterparts using the most popular blue TADF host DPEPO (Fig. 2C and fig. S8) (27). Therefore, tBCzHDPO-based devices can be integrated into the lithium battery–driven portable equipment.

EL spectra of the devices at 4.0 V are identical to PL spectra of the corresponding DMAC-DPS–doped films, with the peaks at 476 nm for tBCzHDPO and tBCzMDPO and 464 nm for tBCzHSPO and tBCzMSPO (Fig. 2D). Significantly different from the other hosts with the stable EL spectra during voltage change, emissions of tBCzHDPO–based devices markedly shift from 480 nm at 3.5 V to 460 nm at 10 V,
corresponding to the Commission Internationale de L’Éclairage (CIE) coordinates of (0.175, 0.303) for blush green and (0.168, 0.192) for deep blue. Consequently, as designed, tBCzHDPO supports its devices with the field-induced allochroic phenomenon (Fig. 2E). It is interesting that the devices show continuous allochroic behavior in direct proportion to the voltage input with emission peaks varying from 480 to 460 nm, with an interval of 4 nm for each increase of 1 V, similar to the solvatochromic characteristics of DMAC-DPS (26). This reveals the gradually transformed and uniformly dispersed low-polarity states of tBCzHDPO molecules in EMLs, which is identical to the solvatochromic behaviors of CT molecules under continuously reduced solvent polarities (30). Furthermore, the operation time to realize the emission change, namely, response time, was in reverse proportion to driving voltage and exponentially dependent on power density (fig. S9). This accurate correspondence between the voltage and emission peaks establishes the basis for purposeful and controllable multilevel color regulation as one of the main challenges for OLEDs.

It is particularly noteworthy that the allochroic process of tBCzHDPO-based TADF devices is irreversible, which is opposite to most of organic and inorganic (31, 32) color-variable LEDs (Fig. 3A). During the first circle of voltage increase from 4 to 8 V, the emission peak successively shifts from 476 to 464 nm, whereas for the second circle, the EL emission becomes invariable with a stable peak wavelength at 464 nm and unchanged profiles identical to those at 8 V in the first circle. Because the device emission color depended on the polarity of the tBCzHDPO matrix, the reversibility of allochroic behavior for these devices should be in accord with that of conformation variation for tBCzHDPO. After transformation, the endo conformer of tBCzHDPO is fixed because of the formation of P=O—H—N IHB, which restrains the reverse transformation, and therefore endows the irreversible allochroic characteristics to its devices. The unique irreversible allochroism clearly differentiates this TADF OLED from RLS- and ETM-based devices, which featured reversible allochroic behaviors.

Consequently, tBCzHDPO makes its DMAC-DPS-doped devices competent for nonvolatile data storage. A six-point OLED matrix was fabricated as a prototype six-bit memory device (Fig. 3B). All bits share one anode line, whereas each bit has an independent cathode line for data read and write. A photometer is used to detect the optical property changes of the devices (scheme S4). In actuality, when applying 8 V as write voltage, it is shown that the contrast of 0 and 1 signals can be easily distinguished by the naked eye. This kind of visible stored data, essentially as a memory integrated with a display, markedly simplifies the information readout process for severe environment and special condition applications, such as military use, personal secret key storage, and inorganic (31, 32) color-variable LEDs (Fig. 3A). During the first circle of voltage increase from 4 to 8 V, the emission peak successively shifts from 476 to 464 nm, whereas for the second circle, the EL emission becomes invariable with a stable peak wavelength at 464 nm and unchanged profiles identical to those at 8 V in the first circle. Because the device emission color depended on the polarity of the tBCzHDPO matrix, the reversibility of allochroic behavior for these devices should be in accord with that of conformation variation for tBCzHDPO. After transformation, the endo conformer of tBCzHDPO is fixed because of the formation of P=O—H—N IHB, which restrains the reverse transformation, and therefore endows the irreversible allochroic characteristics to its devices. The unique irreversible allochroism clearly differentiates this TADF OLED from RLS- and ETM-based devices, which featured reversible allochroic behaviors.

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Device performance
Movie indicating the allochroic process
scheme S1. Synthetic procedure of tBCzHPO.
scheme S2. Device structure and energy level diagram of the devices.
scheme S3. Encryption flow chart of EPWPR-type memory based on irreversible AOLEDs.
fig. S1. Potential variation during conformation transformation of tBCzHPO.
fig. S2. Involved molecular orbitals, contours, and contribution weights of S0
fig. S12. 13C NMR spectrum of tBCzHSPO.
fig. S2. Involved molecular orbitals, contours, and contribution weights of S0
REFERENCES AND NOTES
fig. S1. Potential variation during conformation transformation of tBCzH
scheme S2. Device structure and energy level diagram of the devices.
fig. S9. Correlations between emission peaks, driving voltage, operation time, and power density.
fig. S1. Performance repeatability of the allochroic TADF devices.
fig. S11. 1H nuclear magnetic resonance (NMR) spectrum of tBCzHPSO.
fig. S12. 13C NMR spectrum of tBCzHSPO.
fig. S13. 3P NMR spectrum of tBCzHPSO.
fig. S14. 1H NMR spectrum of tBCzHPO.
fig. S15. 13C NMR spectrum of tBCzHPO.
fig. S16. 31P NMR spectrum of tBCzHPO.
table S1. Physical properties of tBCzHPSO.
table S2. EL performance of DMAC-DPS-based devices.
movie S1. Allochroic TADF device.

REFERENCES AND NOTES
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