Enhancing photovoltaic performance via aggregation dynamics control in fused-ring electron acceptor

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Abstract
A new fused-ring electron acceptor FNIC3 with dynamics controlled aggregation behavior was synthesized. FNIC3 shows strong absorption in 600–900 nm, HOMO/LUMO energy levels of $-5.59/-4.04$ eV, and electron mobility of $1.2 \times 10^{-3}$ cm$^2$ V$^{-1}$ s$^{-1}$. The aggregation of FNIC3 shows strong dependency on film formation time. Prolongation of film formation time promotes the crystallization of FNIC3, leading to improved crystallinity and enlarged aggregate sizes. Aggregation of FNIC3 significantly influences the photovoltaic device parameters. Appropriate aggregation red-shifts the absorption and improves the mobilities of the blend, which contributes to high photocurrent and fill factor thus high power conversion efficiency (PCE). Overaggregation leads to increased nonradiative energy loss and insufficient charge generation, resulting in decreased open-circuit voltage and short-circuit current density. The blends based on PM6:FNIC3 fabricated under proper film formation time exhibit a PCE of 12.3%, higher than those fabricated under short and long film formation time (10.0–10.5%).

KEYWORDS
aggregation dynamics, fused-ring electron acceptor, nonfullerene acceptor, organic solar cell

INTRODUCTION
Organic solar cells (OSCs) have attracted broad attention from both academia and industry owing to some merits, such as low cost, light weight, semitransparency, flexibility, facile fabrication to large-area devices, and potential applications in building-integrated photovoltaics and internet of things. In bulk heterojunction (BHJ) OSCs, proper morphology of active layer is essential for high device performance. The features of optimal morphology include proper domain size, high domain purity, ordered molecular packing, and favorable molecular orientation alignment at both bulk and donor/acceptor interface, ensuring efficient exciton diffusion/splitting and charge transport. The aggregation states of donor and acceptor affect film morphology, and molecular configurations affect the aggregation behaviors of molecules.

For acceptor materials, classical fullerene acceptors possess spherical geometries and thus tend to form crystallites with isotropic electron-transporting capabilities. However, the weak absorption and luminescence of fullerenes lead to low PCEs in OSCs. Nonfullerene acceptors (NFAs) show enhanced absorption and lower aggregation, which contribute to superior PCEs in OSCs. However, the strong absorption of NFAs leads to insufficient charge generation and nonradiative energy loss.

In this work, we report synthesis of a new fused-ring electron acceptor (FNIC3) with dynamics controlled aggregation behavior. FNIC3 shows strong absorption in 600–900 nm, HOMO/LUMO energy levels of $-5.59/-4.04$ eV, and electron mobility of $1.2 \times 10^{-3}$ cm$^2$ V$^{-1}$ s$^{-1}$. The aggregation of FNIC3 shows strong dependency on film formation time, promoting crystallization and enlarged aggregate sizes. Aggregation significantly influences the photovoltaic device parameters, improving the PCE of OSCs. Overaggregation leads to energy loss and insufficient charge generation, resulting in decreased PCE. The blends based on PM6:FNIC3 fabricated under proper film formation time exhibit a PCE of 12.3%, higher than those fabricated under short and long film formation time (10.0–10.5%).
to low contribution to photocurrent and large nonradiative energy loss, respectively, which restrict the power conversion efficiencies (PCEs) of the devices.\cite{9,10} To solve this problem, a variety of new acceptors were developed,\cite{10–13} among which rylene diimides\cite{14,15} and fused-ring electron acceptors (FREAs)\cite{16} are two successful systems. The parent rylene diimides possess planar molecular configuration and thus are highly crystalline and tend to form large crystalline domains (usually micron-scale) in blends, which severely limit exciton dissociation and charge generation. Constructing polymers\cite{17,18} and star-shaped molecules\cite{19–21} are two rational molecular design strategies to prevent intermolecular overaggregation.\cite{22} FREAs, as pioneered by Zhan and coworkers and represented by the star molecule ITIC,\cite{23} possess planar molecular backbones and out-of-plane side chains. The planar configuration of molecular backbone facilitates molecular stacking and electronic coupling, while the out-of-plane orientations of side chains effectively restrain overaggregation. Modifications, such as extending molecular backbone,\cite{24–29} reducing the steric hindrance of side chains\cite{30–34} and introducing noncovalent interactions on backbone\cite{35–42} and/or side chains\cite{43–49}, can enhance intermolecular interactions and thus crystallinity.

Processing techniques also influence the aggregation behaviors of donors and acceptors.\cite{4} Treatments before film formation, such as processing solvents\cite{30,51} solvent additives\cite{52}, and processing temperatures,\cite{53,54}, affect the aggregation dynamics of donors and acceptors during film formation thereby the morphology, while post-treatments like thermal\cite{55} and solvent annealing\cite{56} affect the thermodynamics of donor and acceptor domains and usually lead to enhanced ordering. These extrinsic treatments influence both donor and acceptor. Separate adjustment for the aggregation of donor or acceptor is more desired for morphology control, and can be realized by using molecules with strong aggregation tendency under certain conditions. For example, benzothiadiazole-oligothiophene-based polymer donors show strong temperature-dependent aggregation, and thus the aggregation dynamics of these polymers and accordingly active layer morphology can be controlled by temperature.\cite{57,58} However, there are few examples that reported acceptors with obvious dynamically controlled aggregation behaviors, for example, naphthalene diimide-bithiophene copolymer N2200.\cite{59}

Herein, we synthesized a new two-dimensional (2D)-conjugation fused nonacyclic electron acceptor, FNIC3 (Figure 1A) with halogenated side chains and end groups. Halogen atoms, such as F and Cl, improve intermolecular interactions, thus FNIC3 shows strong crystallinity. Especially, the crystallization of FNIC3 is dynamically controlled, and shows strong dependency on film formation time. With increasing film formation time, FNIC3 molecules aggregate and show red-shifted absorption and higher mobility in blended films, thus improve short-circuit current density ($J_{SC}$) and fill factor (FF) of the devices. Further increasing film formation time leads to overaggregation of FNIC3, which causes more nonradiative energy loss and less charge generation thus lower open-circuit voltage ($V_{OC}$) and $J_{SC}$. The blends formed under short, medium, and long film formation time exhibit PCEs of 10.0%, 12.3%, and 10.5%, respectively.

**RESULTS AND DISCUSSION**

**Synthesis and characterization**

FNIC3 was synthesized as shown in Scheme S1 (Supporting Information). Intermediate 2 was synthesized through Pd-catalyzed Stille coupling reaction between a 2D conjugated benzoi,1,2-b:4,5-b’]dithiophene with fluorinated side chains and ethyl 2-bromothieno[3,2-b]thiophene-3-carboxylate. Grignard reaction on intermediate 2 followed by acid-catalyzed Friedel–Crafts cyclization afforded a fused-ring core 3, which was further formulation through Vilsmeier–Haack reaction yielding aldehyde 4. The final product FNIC3 was obtained via Knoevenagel condensation between aldehyde 4 and 3-(1,1-dicyanomethylene)-5,6-dichloro-1-indanone. All compounds were characterized by mass spectrometry, $^1$H NMR, $^{13}$C NMR, and elemental analysis (Supporting Information). FNIC3 shows good solubility in common organic solvents, such as dichloromethane, chloroform, and chlorobenzene at room temperature.

Single crystal of FNIC3 was grown by liquid diffusion method, where ethanol diffused slowly into chloroform solution at 4°C for 1 week. The crystals were deposited at the bottom of mixed solution and transferred on the loop for single crystal X-ray diffraction measurement at 150 K. The crystal data and structure refinement are listed in Table S1. There are two independent conformational molecules in the crystal structure, which are marked in red and blue in Figures 1B and C. The two conformations exhibit planar and linear A-D-A scaffolds, which tend to form the parallel head-to-tail stacking modes and then tiled into a crossed overlapping pattern. In addition, the average π-π distances in parallel stacking modes are 3.37 and 3.51 Å and in crossed stacking mode is 3.57 Å. FNIC3 shows a tendency to assemble into a multidimensional π-π network, which could be facilely controlled and therefore enhance the photovoltaic performance.

The normalized optical absorption spectra of FNIC3 in dilute chloroform solution (10$^{-6}$ M) and thin film are shown in Figure 1D. In solution, FNIC3 shows strong absorption in 600–800 nm with a high molar absorptivity of 2.2 × 10$^5$ M$^{-1}$ cm$^{-1}$ at the absorption maximum (751 nm). The absorption spectrum of FNIC3 film broadens and the absorption maximum red-shifts 43 nm relative to those in solution, which indicates the aggregation of FNIC3 molecules in film. The optical bandgap of FNIC3 estimated from the absorption edge of the film is 1.40 eV.

Cyclic voltammetry was used to investigate the electrochemical properties of FNIC3 (Figure 1E). FNIC3 shows quasireversible oxidation and irreversible reduction waves with onset oxidation and reduction potentials of 1.23 and –0.32 V, respectively, referring to an Ag/AgCl reference electrode. Assuming the absolute energy level of FeCp$_2$ to be 4.8 eV below vacuum (oxidation potential of FeCp$_2$ to be 0.44 V), the HOMO and LUMO energy levels of FNIC3 are estimated to be –5.59 and –4.04 eV, respectively.

The electron mobility of FNIC3 was measured using the space charge limited current (SCLC) method in electron-only devices with a structure of Al/FNIC3/Al (Figure S1).\cite{60} The electron mobility of FNIC3 is 1.2 × 10$^{-3}$ cm$^2$ V$^{-1}$ s$^{-1}$.
**Photovoltaic properties**

Polymer donor PM6\(^{[61]}\) (Figure 1) shows strong absorption in 300–700 nm, which is complementary with that of FNIC3. The HOMO/LUMO levels of PM6 are \(-5.45/-3.65\) eV, matching with those of FNIC3. BHJ OSCs with structure of indium tin oxide (ITO)/ZnO/PM6:FNIC3/MoO\(_3\)/Ag were fabricated to investigate the photovoltaic property of FNIC3. The device parameters show obvious dependency on film formation time (Figure 2A and Table 1). The blend formed under short film formation time (s-blend), which was spin-coated from a low-boiling-point chloroform solution, shows a PCE of 10.0% with a \(V_{OC}\) of 0.859 V, \(J_{SC}\) of 21.40 mA cm\(^{-2}\), and FF of 0.545. The blend formed under medium film formation time (m-blend), which was spin-coated from high-boiling-point chlorobenzene using a pre-spin-coating process (details described in Supporting Information and Table S2), shows a lower \(V_{OC}\) of 0.775 V but higher \(J_{SC}\) of 22.91 mA cm\(^{-2}\) and significantly higher FF of 0.691, thus a higher PCE of 12.3%. The blend formed under long film formation time (l-blend) was spin-coated from chlorobenzene and exhibits a further improved FF of 0.698 but the lowest \(V_{OC}\) of 0.764 V and \(J_{SC}\) of 19.66 mA cm\(^{-2}\) and a PCE of 10.5%.

The external quantum efficiency (EQE) spectra of the devices based on PM6:FNIC3 under different film formation time are shown in Figure 2B. Arising from the complementary absorption of PM6 and FNIC3, the blends show photoresponse throughout the visible and extending to the NIR region, and the photoresponse in 300–650 nm mainly comes from PM6 and that in 650–900 nm mainly comes from FNIC3. Interestingly, the m-blend and l-blend show broader EQE response with spectral edge at 916 nm, which red-shifts 33 nm relative to that of s-blend (883 nm). According to the absorption spectra of the blended films (Figure S2), PM6
shows almost identical absorption profiles in different blends, while FNIC3 shows red-shifted absorption with the prolongation of film formation time, indicating the expansion of EQE spectra mainly comes from the bathochromic absorption of FNIC3. The broader EQE response region is beneficial to harvesting more light in longer wavelength. Compared with the m-blend, l-blend shows similar spectral profile but lower EQE values, resulting in lower $J_{SC}$. Consequently, the calculated $J_{SC}$ (22.32 mA cm$^{-2}$) of m-blend is higher than those of s-blend (20.92 mA cm$^{-2}$) and l-blend (19.24 mA cm$^{-2}$), all of which are consistent with the $J_{SC}$ measured from $J$-$V$ (errors < 5%, Table 1).

Hole ($\mu_h$) and electron mobilities ($\mu_e$) of the blends were measured by SCLC method with device structures of ITO/PEDOT:PSS/PM6:FNIC3/Au for holes and Al/PM6:FNIC3/Al for electrons (Figure S3 and Table 1). The $\mu_h$ and $\mu_e$ of s-blend are 2.0 $\times$ 10$^{-3}$ and 4.6 $\times$ 10$^{-4}$ cm$^2$ V$^{-1}$ s$^{-1}$, respectively. Increasing film formation time promotes charge transport: m-blend shows improved $\mu_h$ and $\mu_e$ of 2.6 $\times$ 10$^{-3}$ and 9.6 $\times$ 10$^{-4}$ cm$^2$ V$^{-1}$ s$^{-1}$, respectively, and l-blend
shows the highest $\mu_h$ and $\mu_e$ of $4.2 \times 10^{-3}$ and $1.2 \times 10^{-3}$ cm$^2$ V$^{-1}$ s$^{-1}$, respectively. Higher mobilities are beneficial to higher FF.

Charge recombination in the devices was investigated by measuring the $V_{OC}$ and $J_{SC}$ under different light intensities ($P_{\text{light}}$). The slope values of the linear fits of $V_{OC} = \ln P_{\text{light}}$ for s-, m-, and l-blend are 0.99 $k_B T / q$, 1.00 $k_B T / q$, and 1.01 $k_B T / q$ (Figure 2C, $k_B$: Boltzmann constant, $T$: temperature, $q$: elementary charge), suggesting Shockley–Read–HALL recombination is weak and bimolecular recombination is dominant in all of the three blends. The values of $S$ in the equation $J_{SC} \propto P_{\text{light}}^S$, which describes the relationship between $J_{SC}$ and $P_{\text{light}}$, are 0.92 in all blends, suggesting all blends show weak bimolecular recombination (Figure 2D).

The little difference in charge recombination indicates that recombination is not the main reason for different device performance in these blends.

Charge transfer in the devices was investigated by photoluminescence (PL) quenching (Figure S4). PM6 shows two emission peaks at 696 and 798 nm when excited at 555 nm, and the s-, m-, and l-blends show PL quenching of 98%, 98%, and 96% when excited at 555 nm, respectively. FNIC3 shows two emission peaks at 877 and 963 nm when excited at 761 nm, and the s-, m-, and l-blends show PL quenching of 97%, 97%, and 96% when excited at 761 nm, respectively. All blends show obvious PL quenching no matter exciting donor or acceptor, suggesting that both electron transfer and hole transfer are highly efficient.

All blends show weak bimolecular recombination (Figure 2D).

Charge generation and extraction were investigated by measuring the photocurrent density ($J_{ph}$) under different effective voltage ($V_{\text{eff}}$) (Figure 2E). The saturated photocurrent density ($J_{sat}$) is obtained at high $V_{\text{eff}}$ (> 2.5 V), and the ratio of $J_{SC}$ to $J_{sat}$ represents the total efficiency of charge generation and extraction under short-circuit condition. The $J_{sat}$ of s-, m-, and l-blend are 23.56, 24.39, and 21.47 mA cm$^{-2}$, respectively, and $J_{SC}/J_{sat}$ values are 0.908, 0.939, and 0.916, respectively. The $J_{sat}$ of l-blend is ~10% lower than those of s-blend and m-blend, which suggests the total charges generated in l-blend are less. The more efficient charge generation and extraction in m-blend indicated by higher $J_{SC}/J_{sat}$ value is beneficial to higher $J_{SC}$.

The nonradiative energy loss ($E_{loss\text{- non-rad}}$) was characterized by measuring electroluminescence quantum efficiency (EQEEL) of the blend (Figure S5). The EQEEL values of s-, m-, and l-blend are $4.4 \times 10^{-5}$, $2.3 \times 10^{-5}$, and $1.0 \times 10^{-5}$, respectively, from which $E_{loss\text{- non-rad}}$ are calculated to be 0.26, 0.27, and 0.30 eV, respectively, using the equation $E_{loss\text{- non-rad}} = -k_B T \ln(EQEEL)$.

The l-blend shows the largest nonradiative energy loss and thus the lowest $V_{OC}$.

**Film morphology**

The crystallinity and molecular stacking in films were investigated by grazing-incidence wide-angle X-ray scattering (GIWAXS). The PM6 neat films cast under different film formation time show similar scattering features and crystallinity, while the FNIC3 neat film cast under long film formation time shows significantly enhanced crystallinity than that cast under short film formation time, indicating the prolongation of film formation time mainly affects the aggregation behavior of FNIC3 rather than polymer donor (Figure 3 and Table S3). Figure 3 shows the highest $\mu_h$ and $\mu_e$ of $4.2 \times 10^{-3}$ and $1.2 \times 10^{-3}$ cm$^2$ V$^{-1}$ s$^{-1}$, respectively. Higher mobilities are beneficial to higher FF.

Charge recombination in the devices was investigated by measuring the $V_{OC}$ and $J_{SC}$ under different light intensities ($P_{\text{light}}$). The slope values of the linear fits of $V_{OC} = \ln P_{\text{light}}$ for s-, m-, and l-blend are 0.99 $k_B T / q$, 1.00 $k_B T / q$, and 1.01 $k_B T / q$ (Figure 2C).

The nonradiative energy loss ($E_{loss\text{- non-rad}}$) was characterized by measuring electroluminescence quantum efficiency (EQEEL) of the blend (Figure S5). The EQEEL values of s-, m-, and l-blend are $4.4 \times 10^{-5}$, $2.3 \times 10^{-5}$, and $1.0 \times 10^{-5}$, respectively, from which $E_{loss\text{- non-rad}}$ are calculated to be 0.26, 0.27, and 0.30 eV, respectively, using the equation $E_{loss\text{- non-rad}} = -k_B T \ln(EQEEL)$.

The l-blend shows the largest nonradiative energy loss and thus the lowest $V_{OC}$.

Figure 3 (A) 2D GIWAXS patterns of the neat PM6 and FNIC3 cast under different film formation time. (B) Corresponding GIWAXS profiles along the out-of-plane and in-plane directions.
are beneficial to charge transport, which leads to the superior carrier mobility and FF as mentioned above.

The surface morphology of three blends was investigated by atomic force microscopy (AFM). With increasing film formation time, obvious and enlarged aggregates are formed (Figure S6), which are attributed to the crystallization of FNIC3. The aggregation states of FNIC3 significantly affect the absorption, charge transport, charge generation, and hence the photovoltaic performance of the devices. As FNIC3 molecules aggregate, the absorption of the blended films red-shifts, thus the EQE spectra of m-blend and l-blend are broader than that of s-blend, which is beneficial to harvesting lower-energy photons. The crystallinity of m-blend and l-blend is higher than that of s-blend, which is beneficial to charge transport, thus m-blend and l-blend show higher mobilities and FF. Large aggregates in l-blend favor charge transport but reduce the donor/acceptor interface area, which restricts charge generation and leads to low photocurrent and EQE. Therefore, l-blend shows much higher FF but lower $J_{SC}$ relative to s-blend, resulting in a slight enhancement in PCE; m-blend shows the most favorable morphology, which benefits both charge generation and transport, thus exhibits the highest PCE.

The solvent–solute interactions and miscibility between PM6 and FNIC3 are investigated to shed light on the origins of this aggregation behavior of FNIC3. The surface energies of PM6 and FNIC3 calculated from contact angle measurements (Figure S7) using the Owens–Wendt equation $[68]$ are 30.19 and 39.64 mN m$^{-1}$ (Table S4), respectively, from which the Flory–Huggins interaction parameters ($\chi$) can be calculated.$[69]$ According to the Flory–Huggins model, a smaller $\chi$ indicates stronger interactions between two components, namely, better miscibility. The calculated $\chi$ [chlorobenzene, PM6], $\chi$ [chlorobenzene, FNIC3], and $\chi$ [PM6, FNIC3] values are 0.35, 0.85, and 0.98, respectively, which indicates that FNIC3 is less miscible with both solvent and donor, thus shows strong tendency to precipitate from the solvent and form self-aggregates during the film formation process.

AFM images of pristine PM6, pristine FNIC3, and PM6:FNIC3 blended films also confirm this inference (Figure S8). The PM6 films cast from chlorobenzene with the film formation time of 2 s (Figure S8A) and 10 s (Figure S8B) show similar and smooth surfaces, while the FNIC3 film cast from chlorobenzene shows much rougher morphology with micron-scale aggregates, which can be observed under an optical microscope (Figure S8C). In blended films, the FNIC3 aggregates gradually enlarge as the film formation time increases from 2 to 10 s (Figures S8D–F), which can be attributed to the relatively low miscibility between PM6 and FNIC3; the aggregation process of FNIC3 depends on the film formation time, thus can be dynamically controlled to achieve an optimal morphology and thereby improved PCEs.

CONCLUSIONS

In summary, we synthesized FNIC3 whose aggregation states are dynamically controlled and depend on film formation time. FNIC3 neat film shows significantly enhanced...
crystallinity as the film formation time prolongs. Paring with polymer donor PM6, the FNIC3-based blends exhibit distinct morphological, optical, electronic, and photovoltaic properties under different film formation time. Longer film formation time improves the crystallinity of the blend and the sizes of aggregates. The aggregation of FNIC3 redshifts the absorption edge of the blend, thus broadens the photoreponse region. Higher crystallinity is beneficial to charge transport, thereby improves mobilities as well as FF. Overlarge aggregates reduce donor/acceptor interface area and hence suppress exciton dissociation and charge generation, leading to lower EQE and JSC. Overaggregation also increases nonradiative energy loss, leading to decreased Voc.

In conclusion, the blend formed under short film formation time shows a highest Voc; moderate JSC, lowest FF, and lowest PCE of 10.0%; the blend formed under long film formation time shows the highest Voc, OC, moderate JSC, lowest FF, and hence suppress exciton dissociation and charge generation, leading to lower EQE and JSC. Overaggregation also increases nonradiative energy loss, leading to decreased Voc.

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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DATA AVAILABILITY STATEMENT
The data that support the findings of this study are available from the corresponding author upon reasonable request.

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**SUPPORTING INFORMATION**

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