REVIEW

Recent progress of Y-series electron acceptors for organic solar cells

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Abstract
Organic solar cells (OSCs) have entered the era of non-fullerene electron acceptors. Nowadays with the rapid development of non-fullerene electron acceptor materials, Organic photovoltaic show huge potential in challenging conventional photovoltaic technology. Among the emerging non-fullerene electron acceptors, the Y-series acceptors own superior performance. The single-junction devices based Y-series acceptors have achieved power conversion efficiencies of 17∼18%. This paper gives an overview of the recent advances of Y-series electron acceptors. Through discussing the selected molecules, the perspective of molecular design on three parts of the molecules, including the central core, side chain and end group, are spotlighted, and the photoelectric properties, photovoltaic performance and film morphology of various Y-series acceptors materials are also presented. Finally, some molecule design directions will be proposed to further promote the progress of OSCs.

KEYWORDS
non-fullerene acceptors, organic solar cells, Y-series acceptors, Y6

1 | INTRODUCTION

Energy has always been the foundation of human existence. Today in the 21st century, it is still an important project to find renewable and clean energy to replace traditional fossil energy.[1] In a broad sense, all the other clean energy including wind energy, hydropower, and ocean energy are derived from the solar energy.[2] In the terms of solar energy conversion, silicon-based solar cells have become commercial technology to convert solar energy into electric energy. Among the other emerging solar cells, organic solar cells (OSCs) pioneered by Tang in 1986[3] have developed rapidly due to the numerous advantages such as low-cost fabrication, light weight, flexibility and so on.[4] OSCs with sandwich structure contain the electrodes, light active layers and interface layers (Figure 1). Usually, electron donor and acceptor materials cooperate together to realize the absorption and conversion of sunlight. In the beginning of OSCs, electron donor and acceptor materials are one layer each. A milestone breakthrough was the advent of bulk-heterojunction (BHJ) OSCs which blend the donors and acceptors into one single active layer (Figure 1).[5] BHJ OSCs endow excitons with shorter diffusion distance and lower recombination probability, which can improve the performance of OSCs.[6]
Donor materials contain polymer donors and small molecule donors. In polymer solar cells (PSCs), the donor materials are usually D-A type copolymers (D: electron donor units; A: electron acceptor units). The respective polymer donors used in the OSCs are presented in Figure 2. In the first 20 years of the development of acceptor materials, the performance of fullerene acceptors (e.g., Figure 3 PC61BM and PC71BM) have always been on the leading position due to their isotropic transmission and high electron affinity. The study on non-fullerene electron acceptor (NFA) materials has been underway since the beginning of the 21st century. However, it was not until 2015 that the star molecule ITIC was born to break the 20-year monopoly of fullerene acceptors (Figure 3). At this point, OSCs have entered a new era. The power conversion efficiencies (PCEs) of OSCs have been elevated exceed up to 16% through the structure modification of ITIC in about three years. In 2019, Zou proposed A-DA’D-A type fused-ring electron acceptor design strategy, which made big strides in the enhancement of PCEs for OSCs. Once again the emerging A-DA’D-A type Y-series acceptors represented by Y6 have made OSCs enter the fast track of development. Nowadays the PCE of single-junction OSCs devices based on ITIC derivatives reported from 2015 to 2017, BZIC has significant changes in chemical structure, in which a benzothiadiazole (BTZ) was introduced into the middle of cores and cyclopentadienyls were replaced with pyrrole rings. The introduction of electron-deficient BTZ increases intramolecular electronic interactions, while pyrrole rings with strong electron-donating ability also improve charge carrier mobility. Therefore BZIC with six-fused-rings core shows obvious redshift absorption than ITIC with seven-fused-rings core. However the devices based on HFQx-T/BZIC only gave a PCE of 6.3% due to the low short-circuit current density (Jsc) and fill factor (FF) (Table 1). Now it seems that BZIC did not stir many waves in the field at the time. Subsequent reported Y1 and Y2 molecules with extended conjugate length of the central cores showed better performance due to the increase of Jsc and FF when blended with PBDB-T (Table 1). Subsequently Zou et. al. used benzothiadiazole (BT) with stronger electron-withdrawing capability to synthesize Y5 having side chain on the terminal of the central unit. Relative to Y1, Y5 showed narrower optical bandgap because of the elevated intramolecular electronic interactions. Y5 exhibited higher performance when using PBDB-T as donor. The devices gave the PCE of 14.1% with open circuit voltage (Voc) of 0.87 V, Jsc of 22.6 mA cm⁻², and FF of 71.4% (Table 1). Almost at the same time, Zou used 2-(5,6-Difluoro-3-oxo-2,3-dihydro-1H-inden-1-yliden)malononitrile as end groups to obtain Y6, which was blended with PBDB-TF to give the astonishing PCE of 15.7%. So far, Y-series acceptor materials represented by Y6 began to be sought after by the researchers.

2 | THE BIRTH OF THE STAR MOLECULE Y6

Before Y6 came out, Zou and her colleagues have reported one Y6 analogue BZIC (Figure 4) in 2017. Zou pioneered the use of dithieno[3,2-b]pyrrolobenzothiadiazole as cores to construct a new type NFA BZIC. Compared with the ITIC derivatives reported from 2015 to 2017, BZIC has significant changes in chemical structure, in which a benzothiadiazole (BTZ) was introduced into the middle of cores and cyclopentadienyls were replaced with pyrrole rings. The introduction of electron-deficient BTZ increases intramolecular electronic interactions, while pyrrole rings with strong electron-donating ability also improve charge carrier mobility. Therefore BZIC with six-fused-rings core shows obvious redshift absorption than ITIC with seven-fused-rings core. However the devices based on HFQx-T/BZIC only gave a PCE of 6.3% due to the low short-circuit current density (Jsc) and fill factor (FF) (Table 1). Now it seems that BZIC did not stir many waves in the field at the time. Subsequent reported Y1 and Y2 molecules with extended conjugate length of the central cores showed better performance due to the increase of Jsc and FF when blended with PBDB-T (Table 1). Subsequently Zou et. al. used benzothiadiazole (BT) with stronger electron-withdrawing capability to synthesize Y5 having side chain on the terminal of the central unit. Relative to Y1, Y5 showed narrower optical bandgap because of the elevated intramolecular electronic interactions. Y5 exhibited higher performance when using PBDB-T as donor. The devices gave the PCE of 14.1% with open circuit voltage (Voc) of 0.87 V, Jsc of 22.6 mA cm⁻², and FF of 71.4% (Table 1). Almost at the same time, Zou used 2-(5,6-Difluoro-3-oxo-2,3-dihydro-1H-inden-1-yliden)malononitrile as end groups to obtain Y6, which was blended with PBDB-TF to give the astonishing PCE of 15.7%. So far, Y-series acceptor materials represented by Y6 began to be sought after by the researchers.

3 | THE STRUCTURE MODIFICATIONS OF Y6 MOLECULES FOR THE ADVANCE OF OSCS

In terms of the structure modifications of Y6, this work still revolves around core engineering, side chain engineering and end group engineering. The interaction of central cores and end groups can tune the intramolecular charge transfer, light absorption, energy levels, optical bandgap of NFAs, while side chains will affect the solubility, π-π stacking, and aggregation behavior of NFAs. Through the
FIGURE 2  Chemical structures of polymer donors
structure modifications of Y-series acceptors and structure optimization of polymer donors, the single-junction OSCs devices have achieved PCEs of 17~18%.

### 3.1 Core engineering

The deficient aromatic rings are fused with donor units via pyrrole bridges to construct the new D-π-A-π-D type cores.

At first, the attention was focused on the electron-deficient aromatic rings. A series of electronegative motifs have been applied to construct new cores. Besides the early reported BTZ\[^{[13,18]}\] and BT\[^{[20,22]}\] units, Zhu and Lin developed quinoxaline\[^{[23]}\] and benzoselenadiazole\[^{[24]}\] based acceptor molecules one after another. The selected molecules of Y6 \[^{[22]}\], Y11 \[^{[25]}\], AQx-2 \[^{[23b]}\] and Y6Se \[^{[24]}\] are discussed to analyze the effects of the central A’ units on the
TABLE 1  Photoelectric properties of Y-series acceptors and OSCs devices performance based on Y-series acceptors

| NFAs        | HOMO/LUMO  | E_g (eV) | χ_{film} (nm) | Donors     | Voc (eV) | J_sc (mA cm^{-2}) | FF (%) | PCE (%) | Ref. |
|-------------|------------|----------|---------------|------------|----------|-------------------|-------|--------|------|
| BZIC        | -5.42/-3.88 | 1.45     | —             | HFQx-T     | 0.84     | 12.67             | 59    | 6.3    | [19a]|
| Y1          | -5.45/-3.95 | 1.44     | 802           | PBDB-T     | 0.87     | 22.44             | 69.1  | 13.42  | [13] |
| Y2          | -5.43/-4.04 | 1.40     | 827           | PBDB-T     | 0.82     | 23.56             | 69.4  | 13.4   | [13] |
| Y5          | -5.55/-3.87 | 1.38     | 783           | PBDB-T     | 0.88     | 22.8              | 70.2  | 14.1   | [20] |
| Y6          | -5.65/-4.10 | 1.33     | 821           | PBDB-TF    | 0.83     | 25.3              | 74.8  | 15.7   | [22] |
| AQx-1       | -5.59/-3.85 | 1.35     | 820           | PBDB-TF    | 0.89     | 22.18             | 67.14 | 13.31  | [25a]|
| AQx-2       | -5.62/-3.88 | 1.35     | 830           | PBDB-TF    | 0.86     | 25.38             | 76.25 | 16.64  | [23b]|
| Y6Se        | -5.70/-4.15 | 1.32     | 858           | D18        | 0.839    | 27.98             | 75.3  | 17.7   | [24] |
| CH1007      | -5.59/-3.97 | 1.30     | 844           | PBDB-TF    | 0.820    | 27.03             | 72    | 15.96  | [26] |
| BPF-4F      | -5.58/-3.98 | 1.36     | —             | SZ5        | 0.846    | 22.1              | 67.4  | 12.6   | [27] |
| BTP-4F      | -5.59/-4.00 | 1.36     | —             | SZ5        | 0.853    | 24.8              | 79.1  | 16.8   | [27] |
| BPS-4F      | -5.54/-4.00 | 1.29     | —             | SZ5        | 0.822    | 25.4              | 79.9  | 16.3   | [27] |
| N-Cl1       | —          | —        | —             | PBDB-TF    | 0.852    | 21.47             | 70.6  | 12.91  | [30] |
| N3          | —          | —        | —             | PBDB-TF    | 0.837    | 25.81             | 73.9  | 15.98  | [30] |
| N4          | —          | —        | —             | PBDB-TF    | 0.819    | 25.01             | 69.9  | 14.31  | [30] |
| BTP-4F-12   | -5.68/-4.06 | 1.33     | —             | PBDB-TF    | 0.855    | 25.3              | 76    | 16.4   | [31] |
| BTP-4Cl-8   | -5.67/-4.11 | 1.33     | 836           | PBDB-TF    | 0.872    | 25.2              | 74.3  | 16.3   | [32] |
| BTP-4Cl-12  | -5.66/-4.09 | 1.33     | 836           | PBDB-TF    | 0.858    | 25.6              | 77.6  | 17.0   | [32] |
| BTP-4Cl-16  | -5.68/-4.09 | 1.33     | 836           | PBDB-TF    | 0.862    | 24.2              | 74.8  | 15.6   | [32] |
| Y9          | -5.59/-3.78 | 1.36     | 808           | PBDB-T     | 0.90     | 23.28             | 63    | 13.26  | [34] |
| BTP-c7      | -5.62/-4.03 | 1.40     | —             | PBDB-TF    | 0.843    | 24.1              | 73.5  | 14.9   | [35] |
| BTP-e9      | -5.64/-4.05 | 1.40     | —             | PBDB-TF    | 0.839    | 26.2              | 81.1  | 17.8   | [35] |
| BTIC-4Br    | -5.57/-4.11 | 1.33     | 825           | PBDB-TF    | 0.85     | 20.67             | 69.58 | 12.2   | [36d]|
| BTIC-F-m    | -5.42/-3.90 | 1.36     | 805           | PBDB-TF    | 0.92     | 21.41             | 69.09 | 13.61  | [38] |
| BTIC-CI-m   | -5.42/-3.91 | 1.34     | 822           | PBDB-TF    | 0.88     | 21.35             | 69.7  | 13.16  | [38] |
| BTIC-2Br-m  | -5.56/-4.07 | 1.33     | 820           | PBDB-TF    | 0.88     | 25.03             | 73.13 | 16.11  | [36d]|
| BTIC-CF3-m  | -5.45/-3.97 | 1.31     | 839           | PBDB-TF    | 0.85     | 24.89             | 72.32 | 15.3   | [38] |
| BTIC-CF3-y  | -5.45/-3.96 | 1.30     | 837           | PBDB-TF    | 0.85     | 25.19             | 72.82 | 15.59  | [38] |
| Y10         | -5.56/-3.76 | 1.35     | 807           | J11        | 0.89     | 21.21             | 71.55 | 13.46  | [37a]|
| BTIC-BO-4Br | -5.53/-4.09 | 1.35     | 823           | PBDB-TF    | 0.86     | 24.06             | 67.84 | 14.03  | [36d]|
| BTTT-2Cl    | -5.61/-3.98 | 1.36     | 825           | PBDB-TF    | 0.896    | 23.80             | 65.19 | 13.80  | [37b]|
| BTTPC       | -5.47/-3.78 | 1.39     | 798           | PBDB-T     | 0.89     | 22.25             | 73    | 14.51  | [37d]|
| BTTPC-Br    | -5.54/-3.88 | 1.37     | 813           | PBDB-T     | 0.86     | 24.71             | 71    | 15.22  | [37d]|
| BTP-CIBr    | -5.79/-4.00 | 1.38     | 826           | PBDB-TF    | 0.906    | 23.48             | 79    | 16.82  | [39] |
| BTP-CIBr1   | -5.79/-4.03 | 1.33     | 842           | PBDB-TF    | 0.854    | 23.66             | 72.2  | 14.56  | [39] |
| BTP-CIBr2   | -5.80/-4.04 | 1.33     | 843           | PBDB-TF    | 0.845    | 24.97             | 73.6  | 15.54  | [39] |

*Energy values calculated through electrochemical cyclic voltammetry.
*bOptical bandgap calculated based on the onset of film absorption.

do not make huge differences in photoelectric parameters of light absorption and optical bandgap (Table 1). Similar with Y6, both Y11 and AQx-2 were also blended with PBDB-TF to fabricate OSCs devices. After conditions optimization, the devices based on Y11 gave the PCE of 16.54%, while AQx-2 based devices achieved a PCE of 16.64% (Table 1). Liu chose D18 as donor to blend with Y6Se to obtain higher PCE of 17.7% due to the increase of J_sc (Table 1).
Another modification method of the cores is the change of outermost TT (thieno[3,2-b]thiophene) units (Figure 5). On one hand, this part, as a changeful donor motif, can adjust the photoelectric properties of electron acceptors; on the other hand, the interaction between the heteroatoms of D units and the O atoms of end groups can have a significant influence on the molecule geometries and film morphologies of acceptor materials. Much recently, Jen group replaced the outermost thiophene rings with selenophene rings to obtain Y6 analogue CH1007 with wider absorption, narrower optical bandgap and more coplanar conjugate backbone.[26] When blended with PBDB-TF, the devices based on CH1007 yielded a PCE of 15.96% with Voc of 0.82 V, Jsc of 27.03 mA cm⁻² and FF of 72%. Coincidentally, Yan and colleagues systematically investigated the photoelectric properties and photovoltaic performance of BPF-4F, BPT-4F, and BPS-4F containing thieno[3,2-b]furan, thieno[3,2-b]thiophene, and selenopheno[3,2-b]thiophene as the D units respectively. Different O···O, S···O, and Se···O noncovalent interactions endow them with different molecule geometries and packing, which cause the difference of charge transfer properties.[27] BPF-4F is even more twisted than BPT-4F and BPS-4F due to the two O atoms that repulse each other. BPS-4F showed the strongest optical absorption and highest bandgap due to the selenium atoms with strongest electron-donating ability. Regarding the photovoltaic performance, BPT-4F based devices gave the highest PCE of 16.8% with the highest FF value due to suitable molecule packing and high electron mobility, while BPS-4F based devices yielded the PCE of 16.3% with the highest Jsc value because of the widest and strongest absorption of BPS-4F (Table 1).

### 3.2 Side chain engineering

Side chains of Y6 especially the branched alkyl chain on the pyrrole rings are also a big difference in chemical structure in contrast to ITIC type acceptors. In ITIC class molecules, four side chains are oriented in four directions and almost perpendicular to the molecular main plane,[29] while the alkyl chains located on the pyrrole rings of Y6 are partially pointing toward each other (Figure 6). It seems that the alkyl chains connected with nitrogen atoms play a decisive role in solubility of molecules. For example N-C11...
with straight alkyl chains on the N atoms showed poor solubility, while the solubility is significantly enhanced form Y6 to N3 to N4.\textsuperscript{30} Extending the length of the branched side chains is also a good method to improve the solubility. BTP-4F-12 with 2-butoxyctyl chain showed good solubility even in some eco-compatible solvent like o-xylene, 1,2,4-trimethylbenzene and tetrahydrofuran, which also exhibited good photovoltaic performance using these processing solvents.\textsuperscript{31} The relationship between materials morphology and materials performance has always the focus of this field. The blend films of PBDB-TF/N4 showed the larger domain size and predominantly edge-on orientation, while PBDB-TF/N3 blend film showed more face-on orientation and suitable domain size. So the performance of the devices based on N3 is more superior than N4 based devices. The length of the branched alkyl chains can also affect the $\pi$-$\pi$ stacking of material molecules. Among these molecules of BTP-4Cl-8, BTP-4Cl-12, and BTP-4Cl-16, BTP-4Cl-12 has the shortest $\pi$-$\pi$ stacking distance, and showed the best performance whether in spin-coated OSCs or in blade-coated OSCs.\textsuperscript{32}

The other important side chains are located on the $\beta$-positions of outermost thiophene rings of cores (Figure 6). The introduction of this side chains can tune the torsion angles between cores and end groups, molecule packing and stable conformation, thus affect photovoltaic properties and performance.\textsuperscript{33} Y9, whose only differences with Y1 is the undecyl chains, showed higher LUMO energy level and narrower bandgap due to the weak electron-rich undecyl group.\textsuperscript{34} Corresponding devices gave higher Voc. Hou et. al. reported three Y6 analogues BTP-e7, BTP-e9, and BTP-e11 with different lengths of side chains on
the edge of cores. Grazing-incidence wide-angle X-ray scattering (GIWAXS) measurements of neat NFAs films showed that with the increase of the length of the straight chains, the d-space of BTP-e7, BTP-e9, and BTP-e11 increase, which suggest that the side chains on this site affect the interaction of intermolecular end groups. Among the corresponding blend films with PBDB-TF, the strong crystalline property leads to the large domain size of the PBDB-TF/BTP-e7 films. In terms of device results, due to the poor blend morphology, BTP-e7 based devices gave the worst PCE of 14.9% with the lowest Jsc (24.1 mA cm$^{-2}$) and FF (73.5%), while the best performance of BTP-e9 based devices yielded the PCE of 17.8% (Table 1).

3.3 | End group engineering

One main factor for the success of ITIC class NFAs is the use of INCN class end groups (INCN: 2-(3-oxo-2,3-dihydroinden-1-ylidene)malononitrile). Carbonyl group and dicyanovinyl group endow INCN with high electron-withdrawing ability, which can promote the charge transport, widen the sunlight absorption and reduce bandgap. On the other hand, the interaction of intermolecular end groups can promote the π-π stacking. To date, INCN derivatives are still applied into the Y6 and Y6 analogues. Follow the previous pattern, a series of reported halogen-substituted or alkyl-substituted INCN derivatives and thiophene-fused end groups were used to be connected with heptacyclic BT cores to construct various new NFAs (Figure 7). Not so long ago, a new CF$_3$-substituted INCN end groups were developed by He group. The new NFA BTIC-CF$_3$-γ exhibited the smallest dihedral angle than BTIC-F-m, BTIC-Cl-m, and BTIC-CF$_3$-m, which contributes to the intermolecular charge transfer for high Jsc and FF. The devices based on PBDB-TF/BTIC-CF$_3$-γ yielded a PCE of 15.59%, which is slightly higher than that of the isomers mixture BTIC-CF$_3$-m. These results prove...
that the positions of halogen atoms of end groups have an effect on the photovoltaic performance of NFAs. Yan group developed one class of end groups containing both Cl atom and Br atom on different positions to construct three isomers BTP-ClBr, BTP-ClBr1, and BTP-ClBr2.\textsuperscript{[39]} Quantum chemistry calculations were carried out to infer BTP-ClBr with the lowest dipole moments, and GIWAXS measurements concluded that BTP-ClBr owned the lowest crystallization propensity among the isomers. Finally, BTP-ClBr based devices gave the highest PCE of 16.82\% with the highest FF of 79\% due to the balanced hole and electron mobilities, and suitable morphology.

4 | THE MAIN FACTORS FOR THE SUCCESS OF Y-SERIES ACCEPTORS

Undoubtedly, Y-series acceptors have become the state-of-the-art acceptor materials applied into the OSCs. They behaved perfectly whether in PSCs or in all small molecules (ASM) OSCs.\textsuperscript{[40]} To date, the PCEs of PSCs based on Y-series acceptors have achieved exceed 18\%. Ding groups obtained 18.22\% efficiency for binary OSCs based on D18/Y6.\textsuperscript{[15]} Liu and Chen achieved exceed 18\% PCEs using quaternary blends and ternary blends, respectively.\textsuperscript{[41]} In terms of the ASM OSCs, Lu et al. obtained 14.7\% efficiency of binary ASM OSCs based on Y6 through delicate BHJ blend concentration control.\textsuperscript{[40b]} After that, 15.34\% efficiency of ASM OSCs was achieved via the addition of a fullerene additive into the same binary system.\textsuperscript{[42]} Even these backbones have been used to construct polymer semiconductor materials applied into all-polymers OSCs.\textsuperscript{[43]} Behind the great application value, it is particularly important to analyze the main factors in determining the success of Y-series acceptors.

1. Photon energy loss $\Delta E_{\text{loss}}$: relative to inorganic solar cells, the higher $\Delta E_{\text{loss}}$ has always been the sore points for OSCs. The energy losses especially the nonradiative energy losses were reduced in OSCs based on Y-series acceptors, which is beneficial for the high Voc. Sharp absorption tail and high electroluminescence quantum efficiency are the main factors for the reduced radiative energy loss and nonradiative energy loss.

2. Sunlight harvesting: wide and strong absorption is the necessary foundation for the high $J_{\text{sc}}$. The absorption edges for almost all the Y-series acceptors reach near 950 nm, which ensure enough photos for obtaining high photocurrent.

3. Molecular packing: different from the ITIC derivatives with $\pi-\pi$ stacking predominately between end groups, there are two $\pi-\pi$ stacking modes for Y-series acceptor. One is similar with ITIC class NFAs, and another is the $\pi-\pi$ stacking through the interaction of the D’ units, which construct 3D network that facilitate charge transport.

5 | SUMMARY AND OUTLOOK

The PCEs of OSCs based on NFAs have tripled in less than 5 years with the help of the development of ITIC and Y6 class NFAs. The PCE breakthrough of 20\% is within sight. The efficiencies of OSCs based on Y-series acceptors are approaching that of perovskite solar cells, which show the huge potential in future commercial application. While excited, we should also see the existed critical issues in the following: (1) The high cost for the synthesis of Y-series NFAs. Up to 10 steps are needed to synthesize Y6 started from the simplest compounds, which will be an obstacle to commercialization. (2) There have been no major breakthroughs in the structure modification of end groups from ITIC to Y6. To date, almost all the OSCs based on Y-series acceptors achieved the Voc less than 0.90 V, which may be a “pain point” for the further enhancement of PCE. End groups play a key role in tuning the LUMO energy level of acceptors. So it is important of the structure modification of end groups for the Y-series acceptors molecule design. (3) Flexible and large area devices are needed to be developed rapidly for the commercialization. In the future commercial application, flexibility is the great superiority for OSCs that is different from inorganic and perovskite solar cells. Large area devices with high efficiency are the fundamentals of the commercialization.

However, the course of anything cannot depend on a single force. The joint development of materials (donor and acceptor) and device processing has contributed to today’s OSCs. Therefore more effort should be put into materials engineering (active layer and interface layer materials) and device fabrication for a bright future of OSCs.

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CONFLICTS OF INTEREST

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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