Supplementary Information (SI) for Journal of Materials Chemistry A.

Supplementary Information for "OSC-Net: A Multi-fidelity Machine Learning Model for Organic Solar Cell"

Haizhou Yang[1], Adam Wold[2], Junlin Ou[3], Jeromy J. Rech[2], Wei You[4]*, Yi Wang[5]*

[1]*Department of Biomedical Engineering, University of Michigan, Ann Arbor, MI 48109, USA*
[2]*Department of Chemistry, University of North Carolina at Asheville, Asheville, NC 28804, USA*
[3]*Department of Engineering Technology, Middle Tennessee State University, Murfreesboro, TN, 37132, USA*
[4] *Department of Chemistry, University of North Carolina at Chapel Hill, Chapel Hill, NC 27599, USA*
[5]*Department of Mechanical Engineering, University of South Carolina, Columbia, SC 29208, USA*

The experimental dataset was developed using key literature from the organic solar cell community [1–147].

The training and validation loss curves from the pre-training and fine-tuning stage are provided in Figs. S1 and S2.

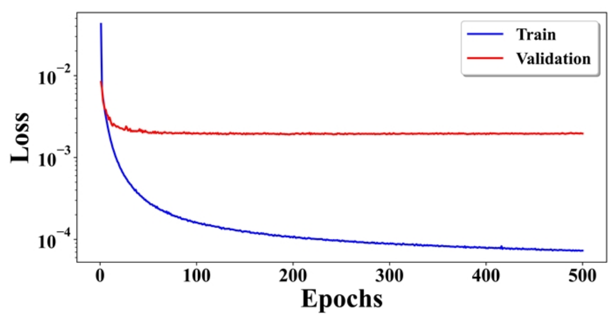

Fig. S1 The training and validation loss curves from the pre-training stage

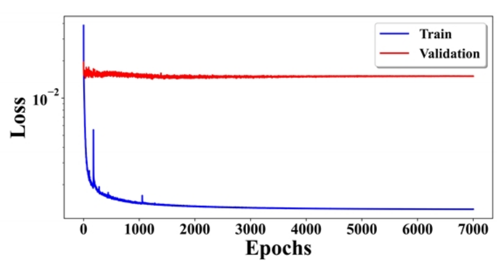

Fig. S2 The training and validation loss curves from the fine-tuning stage

* Corresponding Author.
Yi Wang: Professor, University of South Carolina, yiwang@cec.sc.edu
Wei You: Professor, University of North Carolina at Chapel Hill, wyou@unc.edu

The chemical structures for materials discussed in the main text and highlighted in Table 5 and Table 6 are shown in Figs. S3 (polymer donors) and S4 (small molecule acceptors) below.

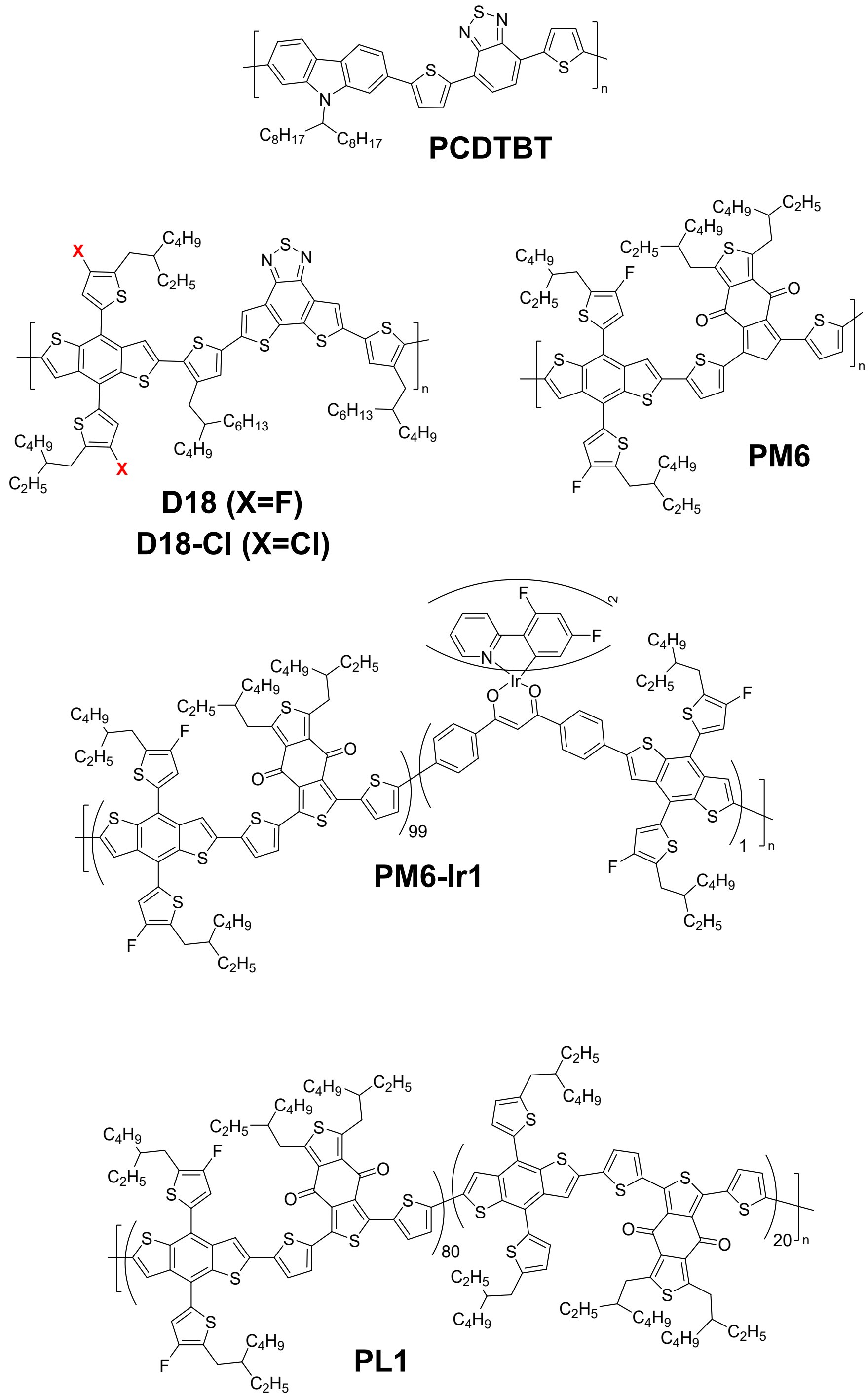

Fig. S3 Polymer Donors

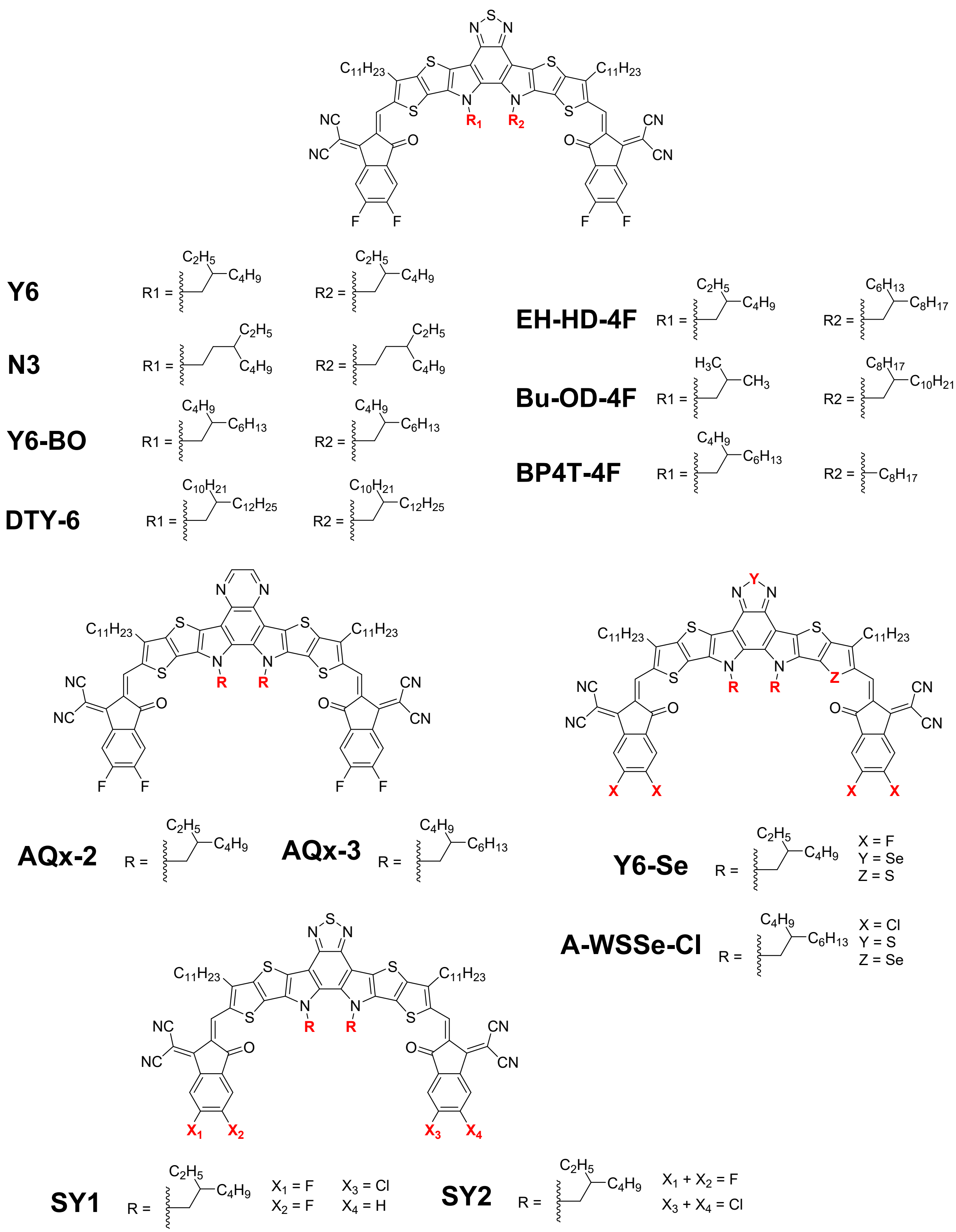

Fig. S4 Small Molecule Acceptors

Reference

1. Chen, W. *et al.* Unsubstituted Benzodithiophene-Based Conjugated Polymers for High-Performance Organic Field-Effect Transistors and Organic Solar Cells. *ACS Appl. Mater. Interfaces* **8**, 19665–19671 (2016).

2. Feng, L. *et al.* Thieno[3,2- b]pyrrolo-Fused Pentacyclic Benzotriazole-Based Acceptor for Efficient Organic Photovoltaics. *ACS Appl. Mater. Interfaces* **9**, 31985–31992 (2017).

3. Zhang, J. *et al.* High-Efficiency Thermal-Annealing-Free Organic Solar Cells Based on an Asymmetric Acceptor with Improved Thermal and Air Stability. *ACS Appl. Mater. Interfaces* **12**, 57271–57280 (2020).

4. Ma, S. *et al.* Heptacyclic S,N-Heteroacene-Based Near-Infrared Nonfullerene Acceptor Enables High-Performance Organic Solar Cells with Small Highest Occupied Molecular Orbital Offsets. *ACS Appl. Mater. Interfaces* **12**, 51776–51784 (2020).

5. Chen, Z. *et al.* Green-Solvent-Processed Conjugated Polymers for Organic Solar Cells: The Impact of Oligoethylene Glycol Side Chains. *ACS Appl. Polym. Mater.* **1**, 804–814 (2019).

6. Zhang, Q. *et al.* Effect of Cyano Substitution on Conjugated Polymers for Bulk Heterojunction Solar Cells. *ACS Appl. Polym. Mater.* **1**, 3313–3322 (2019).

7. Fan, B. *et al.* Tailoring Regioisomeric Structures of π-Conjugated Polymers Containing Monofluorinated π-Bridges for Highly Efficient Polymer Solar Cells. *ACS Energy Lett.* **5**, 2087–2094 (2020).

8. Feng, L.-W. *et al.* Readily Accessible Benzo[d]thiazole Polymers for Nonfullerene Solar Cells with >16% Efficiency and Potential Pitfalls. *ACS Energy Lett.* **5**, 1780–1787 (2020).

9. Fan, B. *et al.* Optimizing Microstructure Morphology and Reducing Electronic Losses in 1 cm 2 Polymer Solar Cells to Achieve Efficiency over 15%. *ACS Energy Lett.* **4**, 2466–2472 (2019).

10. Chai, G. *et al.* Deciphering the Role of Chalcogen-Containing Heterocycles in Nonfullerene Acceptors for Organic Solar Cells. *ACS Energy Lett.* **5**, 3415–3425 (2020).

11. Zhou, C. *et al.* Toward High Efficiency Polymer Solar Cells: Influence of Local Chemical Environment and Morphology. *Adv. Energy Mater.* **7**, (2017).

12. Wang, T. *et al.* Solution-Processed Polymer Solar Cells with over 17% Efficiency Enabled by an Iridium Complexation Approach. *Adv. Energy Mater.* **10**, 1–9 (2020).

13. Chen, Y. *et al.* Side-Chain Engineering on Y-Series Acceptors with Chlorinated End Groups Enables High-Performance Organic Solar Cells. *Adv. Energy Mater.* **11**, 1–10 (2021).

14. Wang, C. *et al.* Low Band Gap Polymer Solar Cells With Minimal Voltage Losses. *Adv. Energy Mater.* **6**, 1–10 (2016).

15. Lee, T. H. *et al.* Investigation of Charge Carrier Behavior in High Performance Ternary Blend Polymer Solar Cells. *Adv. Energy Mater.* **6**, 1–9 (2016).

16. Chen, Y. *et al.* Asymmetric Alkoxy and Alkyl Substitution on Nonfullerene Acceptors Enabling High-Performance Organic Solar Cells. *Adv. Energy Mater.* **11**, 1–10 (2021).

17. Gao, W. *et al.* Asymmetric Acceptors Enabling Organic Solar Cells to Achieve an over 17% Efficiency: Conformation Effects on Regulating Molecular Properties and Suppressing Nonradiative Energy Loss. *Adv. Energy Mater.* **11**, 1–9 (2021).

18. Luo, Z. *et al.* Altering the Positions of Chlorine and Bromine Substitution on the End Group Enables High-Performance Acceptor and Efficient Organic Solar Cells. *Adv. Energy Mater.* **10**, 1–8 (2020).

19. Chang, Y. *et al.* Achieving Efficient Ternary Organic Solar Cells Using Structurally Similar Non-Fullerene Acceptors with Varying Flanking Side Chains. *Adv. Energy Mater.* **11**, 1–9 (2021).

20. He, D. *et al.* A Fused Ring Electron Acceptor with Decacyclic Core Enables over 13.5% Efficiency for Organic Solar Cells. *Adv. Energy Mater.* **8**, 1–7 (2018).

21. Qin, S. *et al.* Non-Halogenated-Solvent Processed and Additive-Free Tandem Organic Solar Cell with Efficiency Reaching 16.67%. *Adv. Funct. Mater.* **31**, 1–10 (2021).

22. Xu, G. *et al.* High-Performance Colorful Semitransparent Polymer Solar Cells with Ultrathin Hybrid-Metal Electrodes and Fine-Tuned Dielectric Mirrors. *Adv. Funct. Mater.*

27, (2017).

23. Mueller, C. J., Singh, C. R., Fried, M., Huettner, S. & Thelakkat, M. High Bulk Electron Mobility Diketopyrrolopyrrole Copolymers with Perfluorothiophene. *Adv. Funct. Mater.* **25**, 2725–2736 (2015).

24. Jo, J. W., Bae, S., Liu, F., Russell, T. P. & Jo, W. H. Comparison of Two D−A Type Polymers with Each Being Fluorinated on D and A Unit for High Performance Solar Cells. *Adv. Funct. Mater.* **25**, 120–125 (2015).

25. Liu, T. *et al.* Asymmetric Acceptors with Fluorine and Chlorine Substitution for Organic Solar Cells toward 16.83% Efficiency. *Adv. Funct. Mater.* **30**, 1–8 (2020).

26. Wan, Q. *et al.* 10.8% Efficiency Polymer Solar Cells Based on PTB7-Th and PC 71 BM via Binary Solvent Additives Treatment. *Adv. Funct. Mater.* **26**, 6635–6640 (2016).

27. Duan, C. *et al.* Wide-Bandgap Benzodithiophene–Benzothiadiazole Copolymers for Highly Efficient Multijunction Polymer Solar Cells. *Adv. Mater.* **27**, 4461–4468 (2015).

28. Xiao, J. *et al.* Surpassing 13% Efficiency for Polythiophene Organic Solar Cells Processed from Nonhalogenated Solvent. *Adv. Mater.* **33**, 1–9 (2021).

29. Zhou, Z. *et al.* Subtle Molecular Tailoring Induces Significant Morphology Optimization Enabling over 16% Efficiency Organic Solar Cells with Efficient Charge Generation. *Adv. Mater.* **32**, 1–8 (2020).

30. Xu, X. *et al.* Single-Junction Polymer Solar Cells with 16.35% Efficiency Enabled by a Platinum(II) Complexation Strategy. *Adv. Mater.* **31**, 1–7 (2019).

31. Cui, Y. *et al.* Single-Junction Organic Photovoltaic Cells with Approaching 18% Efficiency. *Adv. Mater.* **32**, 1–7 (2020).

32. Zhao, F. *et al.* Single-Junction Binary-Blend Nonfullerene Polymer Solar Cells with 12.1% Efficiency. *Adv. Mater.* **29**, 1–7 (2017).

33. Wang, R. *et al.* Rational Tuning of Molecular Interaction and Energy Level Alignment Enables High-Performance Organic Photovoltaics. *Adv. Mater.* **31**, 1–9 (2019).

34. Zhang, S., Qin, Y., Zhu, J. & Hou, J. Over 14% Efficiency in Polymer Solar Cells Enabled by a Chlorinated Polymer Donor. *Adv. Mater.* **30**, 1–7 (2018).

35. Liu, F. *et al.* Organic Solar Cells with 18% Efficiency Enabled by an Alloy Acceptor: A Two-in-One Strategy. *Adv. Mater.* **33**, 1–8 (2021).

36. Gao, Y. *et al.* High Mobility Ambipolar Diketopyrrolopyrrole-Based Conjugated Polymer Synthesized Via Direct Arylation Polycondensation. *Adv. Mater.* **27**, 6753–6759 (2015).

37. Yuan, J. *et al.* Fused Benzothiadiazole: A Building Block for n-Type Organic Acceptor to Achieve High-Performance Organic Solar Cells. *Adv. Mater.* **31**, 1–8 (2019).

38. Liang, Y. *et al.* For the Bright Future—Bulk Heterojunction Polymer Solar Cells with Power Conversion Efficiency of 7.4%. *Adv. Mater.* **22**, 135–138 (2010).

39. Kan, B. *et al.* Fine-Tuning the Energy Levels of a Nonfullerene Small-Molecule Acceptor to Achieve a High Short-Circuit Current and a Power Conversion Efficiency over 12% in Organic Solar Cells. *Adv. Mater.* **30**, 1–8 (2018).

40. Wang, J. *et al.* Enhancing Performance of Nonfullerene Acceptors via Side-Chain Conjugation Strategy. *Adv. Mater.* **29**, 1–7 (2017).

41. Zhang, M. *et al.* Efficient Polymer Solar Cells Based on Benzothiadiazole and Alkylphenyl Substituted Benzodithiophene with a Power Conversion Efficiency over 8%. *Adv. Mater.* **25**, 4944–4949 (2013).

42. Hong, L. *et al.* Eco-Compatible Solvent-Processed Organic Photovoltaic Cells with Over 16% Efficiency. *Adv. Mater.* **31**, 1–7 (2019).

43. Zhou, N. *et al.* Bithiophene Imide and Benzodithiophene Copolymers for Efficient Inverted Polymer Solar Cells. *Adv. Mater.* **24**, 2242–2248 (2012).

44. Li, S. *et al.* Asymmetric Electron Acceptors for High-Efficiency and Low-Energy-Loss Organic Photovoltaics. *Adv. Mater.* **32**, 1–10 (2020).

45. Zhang, M. *et al.* An Easy and Effective Method to Modulate Molecular Energy Level of the Polymer Based on Benzodithiophene for the Application in Polymer Solar Cells. *Adv. Mater.* **26**, 2089–2095 (2014).

46. Sun, C. *et al.* Achieving Fast Charge Separation and Low Nonradiative Recombination Loss by Rational Fluorination for High-Efficiency Polymer Solar Cells. *Adv. Mater.* **31**, 1–8 (2019).

47. Cai, Y. *et al.* A Well-Mixed Phase Formed by Two Compatible Non-Fullerene Acceptors Enables Ternary Organic Solar Cells with Efficiency over 18.6%. *Adv. Mater.* **33**, 1–9 (2021).

48. Zhang, M., Guo, X., Ma, W., Ade, H. & Hou, J. A Polythiophene Derivative with Superior Properties for Practical Application in Polymer Solar Cells. *Adv. Mater.* **26**, 5880–5885 (2014).

49. Chao, P. *et al.* A Benzo[1,2- b :4,5- c ′]Dithiophene-4,8-Dione-Based Polymer Donor Achieving an Efficiency Over 16%. *Adv. Mater.* **32**, 1–8 (2020).

50. Chen, H. *et al.* 17.6%-Efficient Quasiplanar Heterojunction Organic Solar Cells from a Chlorinated 3D Network Acceptor. *Adv. Mater.* **33**, 1–9 (2021).

51. Chen, H. *et al.* 17.1 %-Efficient Eco-Compatible Organic Solar Cells from a Dissymmetric 3D Network Acceptor. *Angew. Chemie - Int. Ed.* **60**, 3238–3246 (2021).

52. Zhou, H. *et al.* Development of Fluorinated Benzothiadiazole as a Structural Unit for a Polymer Solar Cell of 7 % Efficiency. *Angew. Chemie Int. Ed.* **50**, 2995–2998 (2011).

53. Yao, H. *et al.* Design, Synthesis, and Photovoltaic Characterization of a Small Molecular Acceptor with an Ultra-Narrow Band Gap. *Angew. Chemie Int. Ed.* **56**, 3045–3049 (2017).

54. Yang, C. *et al.* A Synergistic Strategy of Manipulating the Number of Selenophene Units and Dissymmetric Central Core of Small Molecular Acceptors Enables Polymer Solar Cells with 17.5 % Efficiency. *Angew. Chemie Int. Ed.* **60**, 19241–19252 (2021).

55. Wang, H. *et al.* Isomerism: Minor Changes in the Bromine Substituent Positioning Lead to Notable Differences in Photovoltaic Performance. *CCS Chem.* **3**, 2591–2601 (2021).

56. Fei, Z. *et al.* Thiophene fluorination to enhance photovoltaic performance in low band gap donor–acceptor polymers. *Chem. Commun.* **48**, 11130 (2012).

57. Wang, Q. *et al.* Effectively Improving Extinction Coefficient of Benzodithiophene and

Benzodithiophenedione-based Photovoltaic Polymer by Grafting Alkylthio Functional Groups. *Chem. – An Asian J.* **11**, 2650–2655 (2016).

58. Wolf, J., Cruciani, F., El Labban, A. & Beaujuge, P. M. Wide Band-Gap 3,4-Difluorothiophene-Based Polymer with 7% Solar Cell Efficiency: An Alternative to P3HT. *Chem. Mater.* **27**, 4184–4187 (2015).

59. Chen, H.-C. *et al.* Prominent Short-Circuit Currents of Fluorinated Quinoxaline-Based Copolymer Solar Cells with a Power Conversion Efficiency of 8.0%. *Chem. Mater.* **24**, 4766–4772 (2012).

60. Kang, T. E., Kim, T., Wang, C., Yoo, S. & Kim, B. J. Poly(benzodithiophene) Homopolymer for High-Performance Polymer Solar Cells with Open-Circuit Voltage of Near 1 V: A Superior Candidate To Substitute for Poly(3-hexylthiophene) as Wide Bandgap Polymer. *Chem. Mater.* **27**, 2653–2658 (2015).

61. Zhang, Z. *et al.* Modification on the Indacenodithieno[3,2- b]thiophene Core to Achieve Higher Current and Reduced Energy Loss for Nonfullerene Solar Cells. *Chem. Mater.* **32**, 1297–1307 (2020).

62. Ye, L., Zhang, S., Zhao, W., Yao, H. & Hou, J. Highly Efficient 2D-Conjugated Benzodithiophene-Based Photovoltaic Polymer with Linear Alkylthio Side Chain. *Chem. Mater.* **26**, 3603–3605 (2014).

63. Jo, J. W. *et al.* Fluorination of Polythiophene Derivatives for High Performance Organic Photovoltaics. *Chem. Mater.* **26**, 4214–4220 (2014).

64. Zhang, Q. *et al.* Fluorinated Thiophene Units Improve Photovoltaic Device Performance of Donor–Acceptor Copolymers. *Chem. Mater.* **29**, 5990–6002 (2017).

65. Bronstein, H. *et al.* Effect of Fluorination on the Properties of a Donor–Acceptor Copolymer for Use in Photovoltaic Cells and Transistors. *Chem. Mater.* **25**, 277–285 (2013).

66. Dai, S. *et al.* Effect of Core Size on Performance of Fused-Ring Electron Acceptors. *Chem. Mater.* **30**, 5390–5396 (2018).

67. Sun, C. *et al.* D–A Copolymer Donor Based on Bithienyl Benzodithiophene D-Unit and

Monoalkoxy Bifluoroquinoxaline A-Unit for High-Performance Polymer Solar Cells. *Chem. Mater.* **32**, 3254–3261 (2020).

68. Shivhare, R. *et al.* Alkyl Branching Position in Diketopyrrolopyrrole Polymers: Interplay between Fibrillar Morphology and Crystallinity and Their Effect on Photogeneration and Recombination in Bulk-Heterojunction Solar Cells. *Chem. Mater.* **30**, 6801–6809 (2018).

69. Li, W., Yan, L., Zhou, H. & You, W. A General Approach toward Electron Deficient Triazole Units to Construct Conjugated Polymers for Solar Cells. *Chem. Mater.* **27**, 6470–6476 (2015).

70. Yang, C. *et al.* Achieving over 10 % Efficiency in Poly(3-hexylthiophene)-Based Organic Solar Cells via Solid Additives. *ChemSusChem* **14**, 3607–3613 (2021).

71. Zhan, L. *et al.* Over 17% efficiency ternary organic solar cells enabled by two non-fullerene acceptors working in an alloy-like model. *Energy Environ. Sci.* **13**, 635–645 (2020).

72. Yang, C. *et al.* Molecular design of a non-fullerene acceptor enables a P3HT-based organic solar cell with 9.46% efficiency. *Energy Environ. Sci.* **13**, 2864–2869 (2020).

73. Jo, J. W. *et al.* Fluorination on both D and A units in D–A type conjugated copolymers based on difluorobithiophene and benzothiadiazole for highly efficient polymer solar cells. *Energy Environ. Sci.* **8**, 2427–2434 (2015).

74. Chai, G. *et al.* Fine-tuning of side-chain orientations on nonfullerene acceptors enables organic solar cells with 17.7% efficiency. *Energy Environ. Sci.* **14**, 3469–3479 (2021).

75. Li, X. *et al.* Effect of the chlorine substitution position of the end-group on intermolecular interactions and photovoltaic performance of small molecule acceptors. *Energy Environ. Sci.* **13**, 5028–5038 (2020).

76. Yang, L., Tumbleston, J. R., Zhou, H., Ade, H. & You, W. Disentangling the impact of side chains and fluorine substituents of conjugated donor polymers on the performance of photovoltaic blends. *Energy Environ. Sci.* **6**, 316–326 (2013).

77. Singh, A. *et al.* Novel fluoride-substituted donor/acceptor polymers containing benzodithiophene and quinoxaline units for use in low–band gap solar cells. *Eur. Polym. J.*

82, 334–346 (2016).

78. Lai, H. *et al.* Trifluoromethylation Enables a 3D Interpenetrated Low-Band-Gap Acceptor for Efficient Organic Solar Cells. *Joule* **4**, 688–700 (2020).

79. Yuan, J. *et al.* Single-Junction Organic Solar Cell with over 15% Efficiency Using Fused-Ring Acceptor with Electron-Deficient Core. *Joule* **3**, 1140–1151 (2019).

80. Dong, S., Jia, T., Zhang, K., Jing, J. & Huang, F. Single-Component Non-halogen Solvent-Processed High-Performance Organic Solar Cell Module with Efficiency over 14%. *Joule* **4**, 2004–2016 (2020).

81. Luo, Z. *et al.* Fine-Tuning Energy Levels via Asymmetric End Groups Enables Polymer Solar Cells with Efficiencies over 17%. *Joule* **4**, 1236–1247 (2020).

82. Ma, Y. *et al.* Efficient Organic Solar Cells from Molecular Orientation Control of M-Series Acceptors. *Joule* **5**, 197–209 (2021).

83. Jiang, K. *et al.* Alkyl Chain Tuning of Small Molecule Acceptors for Efficient Organic Solar Cells. *Joule* **3**, 3020–3033 (2019).

84. Tao, Q. *et al.* Wide bandgap copolymers with vertical benzodithiophene dicarboxylate for high-performance polymer solar cells with an efficiency up to 7.49%. *J. Mater. Chem. A* **4**, 18792–18803 (2016).

85. Qin, R. *et al.* Tuning terminal aromatics of electron acceptors to achieve high-efficiency organic solar cells. *J. Mater. Chem. A* **7**, 27632–27639 (2019).

86. Yu, H. *et al.* Tailoring non-fullerene acceptors using selenium-incorporated heterocycles for organic solar cells with over 16% efficiency. *J. Mater. Chem. A* **8**, 23756–23765 (2020).

87. Li, Z. *et al.* Synergistic effect of fluorination and regio-regularity on the long-term thermal stability of polymer solar cells. *J. Mater. Chem. A* **4**, 18598–18606 (2016).

88. Mo, D. *et al.* Isomeric effects of chlorinated end groups on efficient solar conversion. *J. Mater. Chem. A* **8**, 23955–23964 (2020).

89. Jain, N. *et al.* Interfacial disorder in efficient polymer solar cells: the impact of donor

molecular structure and solvent additives. *J. Mater. Chem. A* **5**, 24749–24757 (2017).

90. Cheung, A. M. H. *et al.* Incorporation of alkylthio side chains on benzothiadiazole-based non-fullerene acceptors enables high-performance organic solar cells with over 16% efficiency. *J. Mater. Chem. A* **8**, 23239–23247 (2020).

91. Wang, Y. *et al.* Engineering the vertical concentration distribution within the polymer:fullerene blends for high performance inverted polymer solar cells. *J. Mater. Chem. A* **5**, 2319–2327 (2017).

92. Gao, W. *et al.* An asymmetrical fused-ring electron acceptor designed by a cross-conceptual strategy achieving 15.6% efficiency. *J. Mater. Chem. A* **8**, 14583–14591 (2020).

93. Chen, Y. *et al.* Alkoxy substitution on IDT-Series and Y-Series non-fullerene acceptors yielding highly efficient organic solar cells. *J. Mater. Chem. A* **9**, 7481–7490 (2021).

94. Guo, B. *et al.* A wide-bandgap conjugated polymer for highly efficient inverted single and tandem polymer solar cells. *J. Mater. Chem. A* **4**, 13251–13258 (2016).

95. Zhu, J. *et al.* Enhancing the performance of a fused-ring electron acceptor via extending benzene to naphthalene. *J. Mater. Chem. C* **6**, 66–71 (2018).

96. Kim, J. *et al.* Synthesis and characterization of highly conjugated side-group-substituted benzo[1,2- b :4,5- b ′]dithiophene-based copolymer for use in organic solar cells. *J. Polym. Sci. Part A Polym. Chem.* **56**, 653–660 (2018).

97. Kranthiraja, K. *et al.* New benzodithiophene- and benzooxadiazole/benzothiadiazole-based donor–acceptor π-conjugated polymers for organic photovoltaics. *J. Polym. Sci. Part A Polym. Chem.* **54**, 2668–2679 (2016).

98. Jin, K., Xiao, Z. & Ding, L. D18, an eximious solar polymer! *J. Semicond.* **42**, 010502 (2021).

99. Qin, J. *et al.* A chlorinated copolymer donor demonstrates a 18.13% power conversion efficiency. *J. Semicond.* **42**, 010501 (2021).

100. Li, G. *et al.* Systematic Merging of Nonfullerene Acceptor π-Extension and Tetrafluorination Strategies Affords Polymer Solar Cells with >16% Efficiency. *J. Am.*

Chem. Soc. **143**, 6123–6139 (2021).

101. Dou, L. *et al.* Systematic Investigation of Benzodithiophene- and Diketopyrrolopyrrole-Based Low-Bandgap Polymers Designed for Single Junction and Tandem Polymer Solar Cells. *J. Am. Chem. Soc.* **134**, 10071–10079 (2012).

102. Son, H. J. *et al.* Synthesis of Fluorinated Polythienothiophene- co -benzodithiophenes and Effect of Fluorination on the Photovoltaic Properties. *J. Am. Chem. Soc.* **133**, 1885–1894 (2011).

103. Zhang, Z. *et al.* Selenium Heterocyclic Electron Acceptor with Small Urbach Energy for As-Cast High-Performance Organic Solar Cells. *J. Am. Chem. Soc.* **142**, 18741–18745 (2020).

104. Kawashima, K. *et al.* Implication of Fluorine Atom on Electronic Properties, Ordering Structures, and Photovoltaic Performance in Naphthobisthiadiazole-Based Semiconducting Polymers. *J. Am. Chem. Soc.* **138**, 10265–10275 (2016).

105. Sun, C. *et al.* High Efficiency Polymer Solar Cells with Efficient Hole Transfer at Zero Highest Occupied Molecular Orbital Offset between Methylated Polymer Donor and Brominated Acceptor. *J. Am. Chem. Soc.* **142**, 1465–1474 (2020).

106. Dai, S. *et al.* Fused Nonacyclic Electron Acceptors for Efficient Polymer Solar Cells. *J. Am. Chem. Soc.* **139**, 1336–1343 (2017).

107. Price, S. C., Stuart, A. C., Yang, L., Zhou, H. & You, W. Fluorine Substituted Conjugated Polymer of Medium Band Gap Yields 7% Efficiency in Polymer−Fullerene Solar Cells. *J. Am. Chem. Soc.* **133**, 4625–4631 (2011).

108. Yao, Z. *et al.* Dithienopicenocarbazole-Based Acceptors for Efficient Organic Solar Cells with Optoelectronic Response Over 1000 nm and an Extremely Low Energy Loss. *J. Am. Chem. Soc.* **140**, 2054–2057 (2018).

109. Li, T. *et al.* Butterfly Effects Arising from Starting Materials in Fused-Ring Electron Acceptors. *J. Am. Chem. Soc.* **142**, 20124–20133 (2020).

110. Lin, F., Jiang, K., Kaminsky, W., Zhu, Z. & Jen, A. K. Y. A Non-fullerene Acceptor with

Enhanced Intermolecular π-Core Interaction for High-Performance Organic Solar Cells. *J. Am. Chem. Soc.* **142**, 15246–15251 (2020).

111. Yao, H. *et al.* 14.7% Efficiency Organic Photovoltaic Cells Enabled by Active Materials with a Large Electrostatic Potential Difference. *J. Am. Chem. Soc.* **141**, 7743–7750 (2019).

112. Liao, X. *et al.* Novel Copolymers Based Tetrafluorobenzene and Difluorobenzothiadiazole for Organic Solar Cells with Prominent Open Circuit Voltage and Stability. *Macromol. Rapid Commun.* **38**, 1600556 (2017).

113. Rech, J. J. *et al.* Utilizing Difluorinated Thiophene Units To Improve the Performance of Polymer Solar Cells. *Macromolecules* **52**, 6523–6532 (2019).

114. Uy, R. L., Yan, L., Li, W. & You, W. Tuning Fluorinated Benzotriazole Polymers through Alkylthio Substitution and Selenophene Incorporation for Bulk Heterojunction Solar Cells. *Macromolecules* **47**, 2289–2295 (2014).

115. Sista, P. *et al.* Synthesis and Electronic Properties of Semiconducting Polymers Containing Benzodithiophene with Alkyl Phenylethynyl Substituents. *Macromolecules* **43**, 8063–8070 (2010).

116. Kularatne, R. S. *et al.* Donor–Acceptor Semiconducting Polymers Containing Benzodithiophene with Bithienyl Substituents. *Macromolecules* **45**, 7855–7862 (2012).

117. Qian, D. *et al.* Design, Application, and Morphology Study of a New Photovoltaic Polymer with Strong Aggregation in Solution State. *Macromolecules* **45**, 9611–9617 (2012).

118. Chang, W.-H. *et al.* A Selenophene Containing Benzodithiophene- alt -thienothiophene Polymer for Additive-Free High Performance Solar Cell. *Macromolecules* **48**, 562–568 (2015).

119. Li, Z. *et al.* Enhancing the performance of the electron acceptor ITIC-Th via tailoring its end groups. *Mater. Chem. Front.* **2**, 537–543 (2018).

120. Li, T. *et al.* A carbon–oxygen-bridged hexacyclic ladder-type building block for low-bandgap nonfullerene acceptors. *Mater. Chem. Front.* **2**, 700–703 (2018).

121. Chai, G. *et al.* Enhanced hindrance from phenyl outer side chains on nonfullerene acceptor

enables unprecedented simultaneous enhancement in organic solar cell performances with 16.7% efficiency. *Nano Energy* **76**, 105087 (2020).

122. Wu, J. *et al.* Carboxylate substituted pyrazine: A simple and low-cost building block for novel wide bandgap polymer donor enables 15.3% efficiency in organic solar cells. *Nano Energy* **82**, 105679 (2021).

123. Cui, Y. *et al.* Organic photovoltaic cell with 17% efficiency and superior processability. *Natl. Sci. Rev.* **7**, 1239–1246 (2020).

124. Ren, J. *et al.* Molecular design revitalizes the low-cost PTV-polymer for highly efficient organic solar cells. *Natl. Sci. Rev.* **8**, (2021).

125. Li, S. *et al.* Unveiling structure-performance relationships from multi-scales in non-fullerene organic photovoltaics. *Nat. Commun.* **12**, 1–11 (2021).

126. Ye, L. *et al.* Unraveling the influence of non-fullerene acceptor molecular packing on photovoltaic performance of organic solar cells. *Nat. Commun.* **11**, 6005 (2020).

127. Li, X. *et al.* Simplified synthetic routes for low cost and high photovoltaic performance n-type organic semiconductor acceptors. *Nat. Commun.* **10**, 519 (2019).

128. Wu, J. *et al.* Random terpolymer based on thiophene-thiazolothiazole unit enabling efficient non-fullerene organic solar cells. *Nat. Commun.* **11**, 4612 (2020).

129. Holliday, S. *et al.* High-efficiency and air-stable P3HT-based polymer solar cells with a new non-fullerene acceptor. *Nat. Commun.* **7**, 11585 (2016).

130. Jia, Z. *et al.* High performance tandem organic solar cells via a strongly infrared-absorbing narrow bandgap acceptor. *Nat. Commun.* **12**, 178 (2021).

131. Yuan, J. *et al.* Enabling low voltage losses and high photocurrent in fullerene-free organic photovoltaics. *Nat. Commun.* **10**, 570 (2019).

132. Sun, C. *et al.* A low cost and high performance polymer donor material for polymer solar cells. *Nat. Commun.* **9**, 743 (2018).

133. Li, C. *et al.* Non-fullerene acceptors with branched side chains and improved molecular

packing to exceed 18% efficiency in organic solar cells. *Nat. Energy* **6**, 605–613 (2021).

134. Liu, S. *et al.* High-efficiency organic solar cells with low non-radiative recombination loss and low energetic disorder. *Nat. Photonics* **14**, 300–305 (2020).

135. Ge, G. *et al.* Design and photovoltaic characterization of dialkylthio benzo[1,2-b:4,5-b′]dithiophene polymers with different accepting units. *Phys. Chem. Chem. Phys.* **17**, 7848–7856 (2015).

136. Wang, Z., Zhao, J., Li, Y. & Peng, Q. Low band-gap copolymers derived from fluorinated isoindigo and dithienosilole: synthesis, properties and photovoltaic applications. *Polym. Chem.* **5**, 4984–4992 (2014).

137. Park, G. E., Kim, H. J., Lee, D. H., Cho, M. J. & Choi, D. H. Regular terpolymers with fluorinated bithiophene units for high-performing photovoltaic cells. *Polym. Chem.* **7**, 5069–5078 (2016).

138. Xiong, J. *et al.* Thiolactone copolymer donor gifts organic solar cells a 16.72% efficiency. *Sci. Bull.* **64**, 1573–1576 (2019).

139. Liu, Q. *et al.* 18% Efficiency organic solar cells. *Sci. Bull.* **65**, 272–275 (2020).

140. Chen, S. *et al.* High-performance polymer solar cells with efficiency over 18% enabled by asymmetric side chain engineering of non-fullerene acceptors. *Sci. China Chem.* **64**, 1192–1199 (2021).

141. Fan, B. *et al.* Achieving over 16% efficiency for single-junction organic solar cells. *Sci. China Chem.* **62**, 746–752 (2019).

142. Luo, Z. *et al.* Conformation-Tuning Effect of Asymmetric Small Molecule Acceptors on Molecular Packing, Interaction, and Photovoltaic Performance. *Small* **16**, 1–9 (2020).

143. Zhang, M. *et al.* Effects of monohalogenated terminal units of non-fullerene acceptors on molecular aggregation and photovoltaic performance. *Sol. Energy* **208**, 866–872 (2020).

144. Shi, Y. *et al.* Optimizing the Charge Carrier and Light Management of Nonfullerene Acceptors for Efficient Organic Solar Cells with Small Nonradiative Energy Losses. *Sol. RRL* **5**, 1–10 (2021).

145. Cho, S.-N. *et al.* Synthesis and characterization of fluorine atom substituted new BDT-based polymers for use in organic solar cells. *Synth. Met.* **210**, 273–281 (2015).

146. Wu, F., Deng, Z., Li, C., Chen, L. & Chen, Y. Structure Evolution of Fluorinated Conjugated Polymers Based on Benzodithiophene and Benzothiadiazole for Photovoltaics. *J. Phys. Chem. C* **119**, 8038–8045 (2015).

147. Nagasawa, S., Al-Naamani, E. & Saeki, A. Computer-Aided Screening of Conjugated Polymers for Organic Solar Cell: Classification by Random Forest. *J. Phys. Chem. Lett.* **9**, 2639–2646 (2018).