Supporting Information for

Benzannulation of Furan: A Strategy for Stable and High-Performance Furan-containing Giant Electron Acceptor with Efficiency Exceeding 20%

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1. Materials and Methods Materials.

Toluene was refluxed and distilled from sodium with benzophenone as an indicator before use. N, N'-Dimethylformamide (DMF), n-Butyllithium (n-BuLi, 1.6 M in THF), and Pd₂(dba)₃ were purchased from Alfa Aesar or J&K Chemical or Derthon (Shenzhen China) Optoelectronic Materials Science Technology Co., Ltd. The synthetic routes to the three BQx-based SMAs were outlined in Scheme S1. Synthetic details of BQx-based compounds and target SMAs were showed as follows. All other reagents and chemicals were purchased from commercial sources and used without further purification¹H and ¹³C NMR spectra were obtained with a Bruker AV-400 MHz NMR spectrometer. Chemical shifts were reported in parts per million (ppm, δ)¹H, and¹³C NMR spectra were referenced to tetramethylsilane (0 ppm) in CDCl₃. Mass spectra were collected on a MALDI Micro MX mass spectrometer, or an API QSTAR XL System.

Device fabrication procedure.

Device Fabrication and Testing The OSCs with conventional structure were fabricated on glass/ITO (indium tin oxide)/PEDOT:PSS (poly(3,4-ethylenedioxythiophene):poly(styrene sulfonate))/active layer/PNDIT-F3N ([(9,9-bis(3'-(N,N-dimethylamino)propyl)-2,7-fluorene)alt-5,5'-bis(2,2'-thiophene)-2, 6-naphthalene-1,4,5,8-tetracaboxylic-N,N'-di(2ethylhexyl)imide])/Ag. The patterned ITO-coated glass substrates were cleaned in detergent, de-ionized water, acetone, and isopropanol sequentially by ultrasonic bath for 30 minutes, respectively, and then dried in an oven at 70 °C overnight. Further UV-Ozone treatment for 20 min was applied before use to improve its work function and clearance. Then the PEDOT:PSS (Heraeus Clevios P VP. AI 4083, filtered at 0.45 µm) was spin-coated onto the cleaned ITOcoated glass substrate at 5500 rpm for 30 s followed by annealing at 150 °C for 15 minutes in air to obtain ~30 nm thick film. The PEDOT:PSS-coated ITO substrates were then transferred into a N₂-filled glove box for further device fabrication. For binary devices, donor:acceptor blends solution with a weight ratio of 1:1.2 were dissolved in the CF, in which the solid concentration is 15.4 mg/mL in total, with 1-chloronaphthalene (0.5% vol) as an additive. For ternary OSCs, PM6:L8-BO:CL25 with a weight ratio of 1:1:0.2 and 15.4 mg/ml total concentration also were dissolved in CF, using DIO (0.3% vol) as additives. Then the blend solution was stirred under 50 °C for 2 hours to mix intensively in a nitrogen-filled glove box.

The blend solution was spin-casted at 3300 rpm for 30 s onto the PEDOT:PSS films followed by a thermal annealing of 100°C for 5 min. For all types of devices, a methanol with 0.5% vol acetic acid blend solution of PNDIT-F3N at a concentration of 0.5 mg mL⁻¹ was spin-coated onto the active layer at 2000 rpm for 30s. The thickness was measured by a profilometer(KLA Tencor D-500). The active layer thickness is ~110 nm measured by Bruker Dektak XT profilometer. A thin layer of PNDIT-F3N (~10 nm) was spin-coated onto the active layer, and Ag electrode (~100 nm) was deposited on top of the electron transfer layer in a thermal evaporator under vacuum of 1×10^{-5} Pa through a shadow mask. The current density-voltage (*J*-*V*) curves of all encapsulated devices were measured using a Keithley 2400 Source Meter under AM 1.5G (100 mW cm⁻²) using an Enlitech solar simulator. The light intensity was calibrated using a standard Si diode with KG5 filter to bring spectral mismatch to unity. Optical microscope (Olympus BX51) was used to define the device area (4.0 mm²). EQEs were measured using an Enlitech QE-S EQE system equipped with a standard Si diode. Monochromatic light was generated from a Enlitech 300W lamp source.

Characterization Methods

Potential surface energy. The optimized molecular structures and the relaxed potential surface energy by bond rotation were calculated by the density functional theory (DFT) method with the Becke three-parameter Lee-Yang-Parr (B3LYP) function and the 6-31G* basis set using a modeling software (Gaussian 09).

UV–visible (UV-Vis) absorption. UV-Vis absorption spectra of films were acquired on a Perkin Elmer Lambda 20 UV/VIS Spectrophotometer. The diluted solutions of BQx-O, BQx-S e and BQx-S were kept at a low concentration of 10⁻⁵ M. All film samples were spin-cast on glass/ITO substrates.

Thermogravimetric (TGA) analysis. Thermogravimetric analysis (TGA) plots were measured with a Discovery series instrument under a nitrogen atmosphere at heating and cooling rates of 10 °C min⁻¹.

Differential scanning calorimetry (DSC) analysis. DSC analysis was conducted on a DSC instrument (DSC Q10) in a temperature range from 25 to 300 °C under N_2 with a heating rate of 10 °C min⁻¹.

Cyclic voltammetry (CV). Cyclic voltammetry was carried out on a CHI610E electrochemical workstation with the three electrodes configuration, using Ag/AgCl as the reference electrode, a Pt plate as the counter electrode, and a glassy carbon as the working electrode. 0.1 mol L⁻¹ tetrabutylammonium hexafluorophosphate in anhydrous acetonitrile was used as the supporting electrolyte. The polymer and small molecules were drop-cast onto the glassy carbon electrode from chloroform solutions (5 mg/mL) to form thin films. Potentials were referenced to the ferrocenium/ferrocene couple by using ferrocene as external standards in acetonitrile solutions. The scan rate is 100 mV s⁻¹. The conversion of reduction/oxidation onsets and LUMO/HOMO energy levels can be described as: equation (2): $E_{LUMO} = -[e(E^{red}-E^{Fc/Fc+})+4.8]$; equation (3): $E_{HOMO} = -[e(E^{ox}-E^{Fc/Fc+})+4.8]$.

Atomic force microscopy (AFM). AFM measurements were performed by using a Scanning Probe MicroscopeDimension 3100 in tapping mode under atmosphere conditions at room temperature. All film samples were spin-cast on glass/ITO substrates.

Grazing incidence wide angle X-ray scattering (GIWAXS). The Grazing-incidence wideangle X-ray scattering (GIWAXS) measurement was carried out with a Xeuss 2.0 SAXS/WAXS laboratory beamline using a Cu X-ray source (8.05 keV, 1.54 Å) and a Pilatus3R 300 K detector. The incidence angle is 0.2°. The samples for GIWAXS measurement were fabricated on the silicon substrates using the same recipe for the devices.

Photoluminescence (PL). The PL measurements were conducted by using FLS1000 equipment. The emission spectra of these three pure NFAs and BQx-O, BQx-Se and BQx-S-based devices were obtained using the same setup used for recording electroluminescence spectra excited by the 514 nm and 785 nm wavelength provided by Xenon lamp. (Detector for NIR 5509 PMT, 600-1700nm).

FTPS-EQE and EL-EQE measurements. FTPS-EQE was measured using a integrated system (PECT-600, Enlitech), where the photocurrent was amplified and modulated by a lock-in instrument. EL-EQE measurements were performed by applying external voltage/current sources through the devices (REPS-Pro, Enlitech). All of the devices were prepared for EL-EQE measurements according to the optimal device fabrication conditions. EL-EQE measurements were carried out from 0 to 1.8 V.

$$\Delta E_{1} = E_{g-q} V_{OC}^{SQ} = E_{g} - k_{B} T \ln \frac{\int_{Eg}^{\infty} \phi AM 1.5 (E) dE}{\int_{Eg}^{\infty} \phi BB (E) dE}$$
$$\Delta E_{2} = k_{B} T \ln \frac{\int_{0}^{\infty} EQE_{PV} (E) \phi BB (E) dE}{\int_{Eg}^{\infty} \phi BB (E) dE}$$

 $\Delta E_1 = E_g - qV_{OC}^{SQ}, \Delta E_1 \text{ is the radiative loss above the bandgap}$ $\Delta E_2 = qV_{OC}^{SQ} - qV_{OC}^{rad}, \Delta E_2 \text{ is the radiative loss below bandgap}$ $\Delta E_3 = qV_{OC}^{rad} - qV_{OC} = -kTln(EQE_{EL}), \Delta E_3 \text{ is the nonradiative loss}$ $V_{OC}^{SQ} \text{ is the maximum } V_{OC} \text{ according to SQ limit, } V_{OC}^{rad} \text{ is the } V_{OC} \text{ when only considering}$ radiative recombination.

Transient photovoltage (TPV) and transient photocurrent (TPC) measurements. In TPV measurements, the devices were placed under background light bias enabled by a focused Quartz Tungsten-Halogen Lamp with an intensity of similar to working devices, *i.e.*, the device voltage matches the open-circuit voltage under solar illumination conditions. Photo-excitations were generated with an 8 ns pulses from a laser system (Oriental Spectra, NLD520). The wavelength for the excitation was tuned to 518 nm with a spectral width of 3 nm. A digital oscilloscope was used to acquire the TPV signal at the open-circuit condition. TPC signals were measured under short-circuit conditions under the same excitation wavelength without

background light bias. The TPV and TPC decay curves were fitted with a single exponential function, and the decay constants were obtained from the fitting.

Charge Carrier Mobility Measurement.

Hole-only diode configuration: glass/ITO/PEDOT:PSS/active layer/MoO₃/Ag. Electron-only diode configuration: glass/ITO/ZnO/active layer/PNDIT-F3N/Ag. The mobility in active layers were determined by fitting the dark current hole/electron-only diodes to the space-charge limited current (SCLC) model. The mobility was determined by the equation:

$J = 9\varepsilon_0 \varepsilon_r \mu_0 V^2 / 8d^3$

where J is current density, μ_0 is the hole or electron mobility, ε_r is the dielectric permittivity of the active layer (generally assumed to be 3 for organic materials), $\varepsilon 0$ is the dielectric permittivity of free space ($\varepsilon_0 = 8.854 \times 10^{-12}$ F m⁻¹), d is the film thickness of active layers, and V is the voltage, which is defined as $V = V_{appl} - V_{bi}$, where V_{appl} is the applied voltage, V_{bi} is the built-in voltage.

Transient absorption spectroscopy. Measurements were performed using a homebuilt experimental setup with an amplified Ti:sapphire laser (Coherent Legend Elite), with pulse duration of 120 fs, centered at 800 nm and at a repetition rate of 1000 Hz. The pump pulses were generated using an optical parametric amplifier (Coherent Opera Solo) and then chopped 500 Hz. The probe beam was traversed a mechanical translation stage, enabling a time delay of up to 2 ns between pump and probe pulses, and then was focused on an Yttrium Aluminium Garnet crystal plate. After passing through the photoexcited sample, the probe pulses were spectrally dispersed using grating and then collected with a silicon line CCD (Hamamatsu S8380) (visible components, 500-1000 nm range) or an InGaAs line CCD (Hamamatsu G11620) (IR components, 800-1600 nm range). The differential transmission signals at various delay times were calculated from the sequential probe shots corresponding to the pump on and off cases as ((T_{pump ON} – T_{pump OFF})/T_{pump OFF}). Measurements were conducted under excitation in fundamental absorption band of the acceptor (800 nm) with an average flux of ~2µJ cm⁻²,

which is close to 1-sun illumination. The decomposition of TA spectra was carried out via softmodelling by multivariate curve resolution-alternating least square (MCR-ALS) method with a singular value decomposition parameters as an initial guess.

In-situ UV-Vis absorption measurements. In-situ UV-Vis absorption measurements were carried out with a DU-100 system. The optical fiber was aligned so that the center of the light is focused on the center of the film. The sampling interval was 5 ms with an integration time of 5 ms per sample point. The average sampling times were adjusted between 7 times to obtain smooth absorption curves.

Multichannel maximum power point tracking (MPPT) measurements. The devices photostabilities were measured by using the solar cell stability test system (PR-SCCS, PURI Materials, China), and PURI2400-8Q as the source meters to record the solar-cells characteristics in an N₂ atmosphere with 45°C, which were conducted under continuous 100 mw/cm2 illumination provided by LED-solar simulators (PR-LEDSUN-8C, PURI Materials, China, with spectrum ranging from 350nm to 900nm). And the results were automatically recorded by the aging test software (PR-SCCS-MPPT, PURI Materials, China). The initial PCEs used in stability studies for BQx-O, BQx-S, and BQx Se-based device were 18.3%, 14.1%, and 12.5%, respectively.

Film-depth-dependent light absorption spectroscopy (FLAS): Film-depth-dependent light absorption spectra were acquired by an in-situ spectrometer (PU100, Shaanxi Puguang Weishi Co. Ltd.) (Shaanxi, China) equipped with a soft plasma-ion source. The power-supply for generating the soft ionic source was 100 W with an input oxygen pressure ~10 Pa. The film surface was incrementally etched by the soft ion source, without damage to the materials underneath the surface, which was in situ monitored by a spectrometer. From the evolution of the spectra and the Beer-Lambert's Law, film-depth-dependent absorption spectra were extracted.

Estimation of glass-transition temperature. UV-vis spectroscopy was used to determine the glass transition temperature (T_g). The absorption spectra of star-shaped acceptor films were measured with increasing temperatures from 25 to 295 °C. For the preparation of films, we used the same processing conditions (i.e., solvent, concentration, and spin coating speed) as those for the OSC fabrication, to precisely correlate the estimated T_g s with those in the OSC device. Then, the deviation metric (DMT) of each absorption spectra was calculated, following the method reported by Harald Ade.^{1,2}

$$DMT = \sum_{\lambda min}^{\lambda max} [IRT(\lambda) - IT(\lambda)]$$

where λ is the wavelength, λ_{max} and λ_{min} are the upper and lower bounds of the optical sweep, respectively, $I_{RT}(\lambda)$ and $I_T(\lambda)$ are the normalized absorption intensities of the as-cast (room temperature) and annealed films, respectively. Then, the T_g is determined to be the point where the two interpolated lines in low- and high-temperature regions intersect.

Synthesis of BQx-O, BQx-Se and BQx-S.

For the synthetic route of compound 2, 4a, 4b, 4c, BQx-O, BQx-Se and BQx-S were synthesized according to the literature method. ³⁻⁵



Scheme S1. The overall synthetic route to BQx-O, BQx-Se and BQx-S.

Synthesis of 2.

To a solution of 1 (400 mg, 0.28 mmol, 1.0 eq.) in 50 ml of tetrahydrofuran was added LiAlH₄ (106 mg, 2.80 mmol, 10 eq.) under an atmosphere of nitrogen. The reaction mixture was kept stirring at 0 °C for 30 minutes. The reaction mixture was heated to 82 °C for 12 hours and then quenched with water. The resulting mixture was extracted with CH_2Cl_2 and the organic layer was washed with brine, dried with anhydrous MgSO₄ and concentrated under a reduced pressure. The crude mixture was used for next step without further purification. To a solution of crude mixture in 40 ml of chloroform was added sequentially 2,3-dichloro-5,6-dicyano-1,4-benzoquinone (DDQ, 73.09 mg, 0.32 mmol, 1.15 eq.) and 4,5-diamino-2-bromobenzonitrile (297 mg, 1.40 mmol, 5.0 eq.). The reaction mixture was kept stirred at room temperature under the atmosphere of nitrogen for 6 hours. The resulting mixture was concentrated under a reduced

pressure and purified by column chromatography on silica gel with hexane/dichloromethane as eluent to afford **2** as a purplish green solid (307 mg, 70%). ¹H NMR (400 MHz, CDCl₃, δ /ppm): 8.76 (s, 1H), 8.66 (s, 1H), 7.04 (s, 2H), 4.67 (d, J = 8.0 Hz, 4H), 2.87-2.82 (m, 4H), 2.17 (s, 2H), 1.88 (s, 4H), 1.48-1.40 (m, 8H), 1.28-1.21 (m, 42H), 1.10-0.92 (m, 24H), 0.88-0.84 (m, 52H), 0.78-0.70 (m, 4H). ¹³C NMR (100 MHz, CDCl₃, δ /ppm):142.81, 141.57, 139.05, 138.45, 137.52, 137.43, 136.44, 136.19, 136.11, 135.76, 131.95, 130.90, 123.04, 122.49, 118.72, 118.58, 118.44, 116.70, 116.06, 115.82, 112.72, 54.88, 38.44, 31.39, 31.36, 31.31, 30.15, 30.07, 29.21, 29.16, 29.07, 29.02, 28.97, 28.94, 28.89, 28.84, 28.79, 28.72, 28.23, 28.19, 25.23, 22.14, 22.12, 22.08, 13.55, 13.53. HRMS (ESI, m/z): [M⁺], calcd. for C₉₅H₁₄₈BrN₅S₄: 1568.4100; found:1568.9847.

Synthesis of 3.

Phosphorus oxychloride (2 mL) was added at 0 °C to anhydrous DMF (10 mL) and the resulting mixture was stirred for 1 h. To a solution of 2 (0.30 g, 0.19 mmol) in 15 ml of 1,2-dichloroethane was added the mixture dropwise. After stirring for additional 30 minutes at 0 °C, the mixture was then heated to 85 °C for overnight and then quenched by 20 ml of 1 M NaOH solution. The reaction mixture was cooled and partitioned between dichloromethane and water. The aqueous layer was extracted with dichloromethane twice. The combined organic layer was washed with brine, dried with anhydrous MgSO₄, and concentrated under reduced pressure. The crude mixture was purified by column chromatography on silica gel with hexane/dichloromethane as eluent to afford **3** as an orange solid. (276 mg, 89%). ¹H NMR (400 MHz, CDCl₃, δ /ppm): 10.16 (s, 2H), 8.82 (s, 1H), 8.75 (s, 1H), 4.68 (d, J = 8.0 Hz, 4H), 3.26-3.21 (m, 4H), 2.15-2.08 (m, 2H), 1.98-1.92 (m, 4H), 1.53-1.50 (m, 4H), 1.44-1.39 (m, 4H), 1.33-1.20 (m, 42H), 1.11-1.05 (m, 24H), 0.97-0.80 (m, 52H), 0.79-0.67 (m, 4H). ¹³C NMR (100 MHz, CDCl₃, δ/ppm):181.04, 145.85, 144.17, 144.10, 141.21, 138.02, 137.30, 136.60, 136.17, 135.99, 135.31, 131.99, 131.52, 131.04, 128.48, 127.09, 126.85, 119.51, 116.40, 116.14, 115.73, 113.53, 55.08, 52.80, 38.91, 31.30, 31.27, 31.23, 30.09, 30.01, 29.90, 29.86, 29.43, 29.28, 29.10, 29.06, 28.99, 28.96, 28.93, 28.85, 28.75, 28.70, 28.65, 27.48, 25.34, 22.07, 22.04, 22.00, 13.48, 13.45, 13.43. HRMS (ESI, m/z): [M⁺], calcd. for C₉₇H₁₄₈BrN₅O₂S₄: 1624.4300; found:1624.9761.

Synthesis of 5a

To a mixture of **3** (531mg, 0.327 mmol), **4a** (37 mg, 0.054 mmol), $Pd_2(dba)_3$ (2.5 mg, 0.0027 mmol), and P-(*o*-tolyl)₃ (3.3 mg, 0.0108 mmol) was added 40 ml of toluene under an atmosphere of nitrogen. The resulting mixture was refluxed for 16 hours. The reaction mixture was cooled and partitioned between dichloromethane and water. The aqueous layer was extracted with dichloromethane twice. The combined organic layer was washed with brine, dried with anhydrous MgSO₄, and concentrated under reduced pressure. The crude mixture was purified by column chromatography on silica gel with hexane/dichloromethane as an eluent to afford **5a** as a brown solid (216 mg, 83% yield).

5b and 5c were synthesized by a similar method with a yield of 80% and 81% respectively.

5a: ¹H NMR (400 MHz, CDCl₃, δ /ppm): 10.20 (s, 6H), 9.34 (s, 3H), 9.08 (s, 3H), 8.59 (s, 3H), 4.79-4.66 (m, 12H), 3.37-3.24 (m, 12H), 2.18-2.08 (m, 6H), 2.04-1.96 (m, 12H), 1.57-1.52 (m, 12H), 1.46-1.45 (m, 12H), 1.33-1.14 (m, 246H), 0.98-0.86 (m, 108H), 0.72-0.61 (m, 12H). ¹³C NMR (100 MHz, CDCl₃, δ /ppm):201.95, 181.13, 181.08, 178.18, 146.22, 143.80, 142.28, 139.92, 138.79, 136.49, 132.49, 132.24, 128.69, 127.57, 116.91, 111.23, 111.22, 54.99, 38.49, 31.32, 31.14, 30.16, 29.86, 29.77, 29.05, 28.94, 28.76, 28.69, 27.64, 24.94, 22.08, 21.98, 13.51, 13.40. HRMS (ESI, m/z): [M⁺], calcd. for C₃₀₃H₄₄₇N₁₅O₉S₁₂: 4828.7200; found:4829.1716.

5b: ¹H NMR (400 MHz, CDCl₃, δ/ppm): 10.17 (d, *J* = 4.0 Hz, 6H), 9.09 (s, 3H), 8.91 (s, 3H), 8.63 (s, 3H), 4.77-4.66 (m, 12H), 3.35-3.21 (m, 12H), 2.20-2.15 (m, 6H), 2.08-1.92 (m, 12H), 1.58-1.41 (m, 12H), 1.34-1.10 (m, 246H), 1.00-0.86 (m, 108H), 0.77-0.67 (m, 24H). ¹³C NMR (100 MHz, CDCl₃, δ/ppm):210.00, 202.84, 199.46, 188.26, 183.21, 181.16, 170.51, 166.65, 163.88, 160.04, 146.38, 143.88, 136.42, 128.78, 127.65, 54.90, 31.32, 31.29, 31.23, 30.81, 29.98, 29.88, 29.56, 29.17, 29.09, 29.04, 28.97, 28.92, 28.89, 28.85, 28.76, 28.67, 24.99, 22.08, 22.00, 13.51, 13.43. HRMS (ESI, m/z): [M⁺], calcd. for C₃₀₃H₄₄₇N₁₅O₆S₁₅: 4876.9100; found:4870.4680.

5c: ¹H NMR (400 MHz, CDCl₃, δ/ppm): 10.18 (d, *J* = 4.0 Hz, 6H), 9.07 (s, 3H), 8.81 (s, 3H), 8.65 (s, 3H), 4.78-4.64 (m, 12H), 3.35-3.18 (m, 12H), 2.22-2.06 (s, 6H), 2.04-1.94 (s, 12H),

1.53-1.42 (m, 12H), 1.28-1.12 (m, 234H), 0.97-0.86 (m, 132H), 0.78-0.59 (m, 12H). ¹³C NMR (100 MHz, CDCl₃, δ /ppm):181.08, 146.16, 143.86, 138.88, 136.49, 136.39, 128.70, 127.69, 116.89, 55.06, 38.43, 31.32, 30.02, 29.18, 29.04, 28.76, 28.67, 27.59, 24.97, 22.08, 13.51, 13.43. HRMS (ESI, m/z): [M⁺], calcd. for C₃₀₃H₄₄₇N₁₅O₆S₁₂Se₃: 5017.6400; found:5017.9247.

Synthesis of BQx-O, BQx-Se and BQx-S.

To a mixture of **5a** (220 mg, 0.046 mmol), 5,6-difluoro-3-(dicyanomethylidene)-1-one (106 mg, 0.46 mmol), and pyridine (1.5 mL) was added 15 ml of chloroform. The resulting mixture was refluxed for 2 hours. The resulting mixture was concentrated and directly purified by column chromatography on silica gel with hexane/dichloromethane as an eluent to afford **BQx-O** as a black solid (188 mg, 67%). **BQx-S** and **BQx-Se** were synthesized by a similar method with a yield of 63% and 65% respectively.

BQx-O: ¹H NMR (400 MHz, CDCl₃, δ/ppm): 9.0-8.40 (m, 12H), 8.36-8.06 (m, 3H), 7.91-7.64 (m, 12H), 5.26-4.91 (m, 24H), 2.87-2.33 (m, 12H), 1.94-0.95 (m, 372H), 0.75-0.46 (s, 24H). HRMS (ESI, m/z): [M⁺], calcd. for C₃₇₇H₄₅₉F₁₀N₂₉O₉S₁₂: 6115.7200; found:6115.0190.

BQx-S: ¹H NMR (400 MHz, CDCl₃, δ /ppm): 9.34-9.27 (m, 3H), 9.19-8.99 (m, 3H), 8.83-8.54 (m, 9H), 8.09-7.59 (m, 12H), 5.29-4.74 (m, 24H), 3.56-2.93 (m, 12H), 2.47-1.95 (m, 30H), 1.48-0.84 (m, 342H), 0.74-0.30 (m, 24H). HRMS (ESI, m/z): [M⁺], calcd. for C₃₇₇H₄₅₉F₁₀N₂₉O₆S₁₅: 6163.9000; found:6163.9280.

BQx-Se: ¹H NMR (400 MHz, CDCl₃, δ/ppm): 8.83-8.55 (m, 9H), 8.50-8.39 (m, 3H), 8.33-8.30 (m, 3H), 8.11-7.66 (m, 12H), 5.39-4.65 (m, 24H), 3.56-2.85 (m, 12H), 2.59-2.22 (m, 6H), 2.06-1.95 (m, 12H), 1.58-1.04 (m, 246H), 0.89-0.58 (m, 132H). HRMS (ESI, m/z): [M⁺], calcd. for C₃₇₇H₄₅₉F₁₀N₂₉O₆S₁₂Se₃: 6304.6300; found:6304.1220.

3. Supplementary Figures (S1-S37).

3.1 Photophysical properties of BQx-O, BQx-Se and BQx-S.



Figure. S1. (a) Thermogravimetric analysis (TGA) curves of BQx-O, BQx-Se, and BQx-S. (b) DSC curves (2^{nd} heating cycles). (c) Normalized UV-vis absorption spectra of three SMAs of thin films. (d) CV curves of BQx-O, BQx-Se, and BQx-S films. (e) J_{SC} vs PCE (inside) and FF vs PCE (outside) for binary OSCs with giant acceptors. (f) Current density-voltage (J-V) curves of the D18:BQx-O binary devices.



Figure. S2. Theoretical density distribution for the frontier molecular orbits of BQx-O, BQx-Se and BQx-S.



Figure. S3. Film UV-Vis absorption spectra of (a) BQx-O, (b) BQx-Se, (c) BQx-S (d) and BQx-CN depending on annealing temperatures. (e) Plots of the DMT of BQx-CN films as a function of annealing temperature. (f) UV/vis absorption spectra of BQx-O (red line), BQx-O (black line, thin film after one year of storage under ambient conditions (room temperature, exposure to light and air), BQx-S (grey line) and BQx-Se (blue line) in thin film. (g) Pure thin film and (h) blend film UV-Vis absorption spectra of BQx-O, BQx-Se, BQx-S after 100 hours of light exposure.



Figure. S4. (a-c) Semilogarithmic plots of normalized EL, measured EQE, and EQE calculated by FTPS (EQE_{FTPS}) as a function of energy for devices based on (a), PM6:BQx-O, (b), PM6:BQx-Se and (c), PM6:BQx-S. (d) Normalized FTPS-EQE based on the PM6:BQx-O, PM6:BQx-Se and PM6:BQx-S. (e) The optical gaps in these devices were measured and derived from EQE. (f) EQE_{EL} spectra of the PM6:BQx-O, PM6:BQx-Se and PM6:BQx-S.



Figure. S5. (a) Histograms of the μ_h and μ_e of the OSCs based on the PM6:BQx-O, PM6:BQx-S and PM6:BQx-S. (b-c) The hole mobility and electron mobility of PM6:BQx-O, PM6:BQx-S e and PM6:BQx-S films.



Figure. S6. (a) Plots of J_{ph} versus V_{eff} . Dependence of (b) J_{SC} and (c) V_{OC} on the Plight of optimized OSCs. (d) Current density-voltage (*J-V*) curves of the BQx-O-based ternary devices (e) EQE spectra of the optimized ternary devices.



Figure S7. The time-resolved UV-Vis absorption spectra of the (a) BQx-O-, (b) BQx-Se- and (c) BQx-S-based blends. In-situ UV-Vis absorption line profiles of (d) BQx-O -, (e) BQx-Se - and (f) BQx-S -based blends.



Figure. S8. (a-c) AFM phase images.



Figure. S9. (a-c) Film-depth-dependent absorption spectra in blend films. (d) Exciton generation rate G profiles simulated from film-depth-dependent absorption spectra with an optical transfer matrix model.



Figure S10. (a-c) The compositional distribution as a function of film depth in blend films. (d-f) Simulated exciton generation contours as a function of film depth.



Figure. S11. Photoluminescence (PL) spectra excited at (a) 514 nm and (b) 785 nm of BQx-O, BQx-Se and BQx-S-based blend films.



Figure S12. (a) Normalized photoluminescence spectra and (b) proposed scheme of recombination pathways based on PL data.



Figure S13. TA spectroscopy of pure trimer films. (a, b) TA spectra collected at early (0.1-1ps) and late time (100-200ps) after photoexcitation. TA kinetics probed at (c) GSB range (830-850 nm) and (d) at long-wavelength shoulder of PA band (950-970 nm for trimer and 970-990 nm for monomer). (e)TA spectra collected at late time (100-200ps) after photoexcitation of blend trimer films.



Figure S14. (a) Normalized TA kinetics of pure acceptor films probed in GSB and (b) longwavelength shoulder of PA, shown in absolute values in Figures S11c and 11d. (c) Normalized TA kinetics of blended films probed in acceptor and donor (d) GSB, shown in absolute values in Figures 6h and 6i of the main text.



Figure S15. (a) Normalized TA kinetics of pure acceptor films probed in GSB and (b) longwavelength shoulder of PA, shown in absolute values in Figures S11c and 11d. (c) Normalized TA kinetics of blended films probed in acceptor and donor (d) GSB, shown in absolute values in Figures 6h and 6i of the main text.

3.2 Photovoltaic device performance.



Figure. S16. Structure of compound PM6, D18 and L8-BO.



Figure. S18. ¹³C NMR spectrum of compound 2.



Figure. S19. HRMS of compound 2



Figure. S20. ¹H NMR of compound 3.



Figure. S21. ¹³C NMR spectrum of compound 3.



Figure. S22. HRMS of compound 3.



Figure. S24. ¹³C NMR spectrum of compound 5a.



Figure. S25. HRMS of compound 5a.



Figure. S26. ¹H NMR spectrum of 5b.



Figure. S27. ¹³C NMR spectrum of 5b.



Figure. S28. HRMS of 5b.



Figure. S29. ¹H NMR spectrum of compound 5c.



Figure. S30. ¹³C NMR spectrum of compound 5c.



Figure. S31. HRMS of compound 5c.



Figure. S32. ¹H NMR spectrum of BQx-O.



Figure. S34. ¹H NMR spectrum of BQx-S.









Figure. S37. HRMS of BQx-Se.

4. Supplementary Tables S1-S8.

SMAs	<i>T</i> _d ^{<i>a</i>} (°C)	<i>T</i> _m ^b (°C)	$\lambda_{\max, sol} c$ (nm)	$\lambda_{ m max, film} d$ (nm)	$\lambda_{\text{onset}}^{e}$ (nm)	Eg ^{opt f} (eV)	E _{HOMO} ^{g/h} (eV)	E _{LUMO} ^{g/h} (eV)	E _{HOMO/LUMO} gap CV/DFT
BQx-O	317.2	275.4	779	810	868	1.43	-5.72/-5.62	-3.66/-3.60	2.06/2.02
BQx-Se	315.1	270.1	743	793	859	1.45	-5.67/-5.67	-3.59/-3.63	2.08/2.04
BQx-S	311.3	272.3	745	796	863	1.44	-5.69/-5.63	-3.63/-3.63	2.06/2.00

 Table S1. The optical and electrochemical parameters of SMAs in this work.

^{*a*} The 5% weight-loss temperatures measured by TGA. ^{*b*} Melting temperature measured by DSC. ^{*c*} Absorption maximum peak of solution. ^{*d*} Absorption maximum peak of thin film. ^{*e*} Absorption onset of the $\lambda_{\text{max,film}}$. ^{*f*} Optical bandgap estimated from the absorption onset of the thin film: $E_{g}^{\text{opt}} = 1240/\lambda_{\text{onset}}$. ^{*g*} Calculated according to the equation: $E_{\text{HOMO/LUMO}} = -e(E_{\text{ox}/\text{red}} + 4.80)$ (eV). ^{*h*} Calculated from the DFT.

 Table S2. The binary photovoltaic properties of furan containing acceptors recorded in this outlook.

Active laver	V _{oc}	J_{sc}	FF	PCE	Ref
	(V)	$(mA cm^{-2})$	(%)	(%)	
PM6: BQx-O	0.912	26.25	77.78	18.62	This work
PBBDTTT-C-T:DC-TDT2T	0.900	8.33	52.30	3.93	6
PBBDTTT-C-T:DC-TDT2F	0.940	5.00	47.80	2.26	7
P3HT: II	0.600	7.90	33.00	1.20	8
PTB7-Th:FRd ₂	0.830	15.70	72.30	9.40	9
PTB7-Th:FRdCN ₂	0.930	16.00	71.90	10.70	10
PTB7-Th:FPDI-F	0.920	8.71	40.00	3.20	11
PBDB-T:FDI ₂	1.060	0.55	25.60	0.15	12
PBDB-T:F-FDI ₂	1.040	9.88	61.60	6.33	12
PM6:IDF-IC	0.900	14.55	59.00	7.80	13
PM6: IDF-4F	0.730	17.69	61.00	7.81	13
PM6: M8	0.830	8.36	60.50	4.21	14
PM6:BFHIC	0.970	15.90	55.80	8.70	15

PM6:BFHIC-4F	0.870	22.80	67.70	13.40	15
PBDB-T:IDTF-4F	0.840	21.30	62.00	11.50	16
PBDB-T:IDTMeF-4F	0.850	23.40	64.00	12.90	16
PBDB-T:IDFF-4F	0.780	11.40	57.00	5.50	16
PBDB-T:IDFT-4F	0.800	12.00	53.00	5.60	16
	0.800	12.00	55.00	5.00	

		-	-		
A (* 1	V_{oc}	J_{sc}	FF	PCE	Ref
Active layer	(V)	$(mA cm^{-2})$	(%)	(%)	iter
PM6: BQx-O	0.912	26.25	77.78	18.62	This work
D18-Cl-B:4A-DFIC	0.905	22.47	77.40	15.76	17
PM6:CH8	0.889	19.70	53.50	9.37	18
РМ6: СН8-0	0.936	22.61	72.10	15.26	19
PM6:CH8-1	0.923	24.89	74.20	17.05	19
PM6:CH8-2	0.928	24.24	74.90	16.84	19
D18: DP-BTP	0.960	22.73	69.10	15.08	20
PM6:CH25	0.840	4.82	55.40	2.24	21
PM6:CH26	0.920	22.98	72.70	15.41	21
D18: DT-6IC	0.961	22.31	73.49	15.66	22
PM6:CH8-3	0.915	24.44	77.00	17.22	23
PM6:CH8-4	0.894	26.05	75.50	17.58	23
PM6:CH8-5	0.902	24.75	75.20	16.79	23
PM6:HDY-m-TAT	0.915	23.22	67.80	14.42	24
PM6:FDY-m-TAT	0.911	26.47	74.70	18.07	24
РМ6: НDY-<i>о</i>-ТАТ	0.933	23.07	70.70	15.23	24
РМ6: FDY-<i>о</i>-TAT	0.914	25.14	73.60	16.91	24
PM1:G-DF	1.015	11.27	53.30	6.10	25
PM6:CH-D1	0.949	23.32	73.20	16.62	26
PM6:Dimer-QX	0.933	22.57	69.26	14.59	27
PM6:Dimer-2CF	0.900	26.39	80.03	19.02	27
PM6: 3BTT6F	0.938	24.21	77.17	17.52	28
PM6: 3BTT6Cl	0.903	23.72	74.36	15.92	28
PM6: 3BY	0.969	23.92	76.59	17.75	29
РМ6:3QY	0.951	26.36	76.86	19.27	29
PM6:G-Trimer	0.896	26.75	79.30	19.01	30

 Table S3. The binary photovoltaic properties of giant acceptors recorded in this outlook.

PM6:TBT	0.940	24.63	78.07	18.04	31
PM6:Tri-BT	0.933	25.49	74.90	17.81	32
PM6:Tri-Qx	0.935	25.28	77.50	18.33	32
PM6:TQT	0.944	25.78	76.10	18.52	32
PM6: Tri-Y6-OD	0.916	25.30	77.80	18.03	33
PM6:tBTICr-BO	0.900	21.25	68.76	13.16	34
PM6: BTY	0.864	27.06	78.01	18.24	35
PM6:TYT	0.964	25.07	75.00	18.15	36
PM6: TYT-S	0.964	25.18	77.00	18.61	37

Table S4. Summary of device parameters based BQx-O of OSCs.

		J_{sc}	$J_{\rm SC},_{\rm cal}{}^a({\rm mA~cm^2})$	FF	PCE
Active layer	$V_{oc}(\mathbf{V})$	$(mA cm^{-2})$		(%)	(%)
D18:BOx-O	0.965 ± 0.002	21.71 ± 0.1		56.376 ± 0.008	11.59 ± 0.3
21012 (.1.0	0.967	21.81		56.38	11.89
PM6:L8-BO	$0.887 {\pm} 0.003$	26.43±0.20	25.57	77.10±0.51	18.20±0.21
1110120 20	0.890	26.63		77.61	18.41
D19.1 9 DO	0.922 ± 0.002	26.33±0.20	25.28	79.01±0.50	19.16±0.20
D19:19-BO	0.924	26.35		79.51	19.36

Table S5. Detailed E_{loss} parameters of the OSCs based on BQx-O, BQx-Se and BQx-S.

	E_{g}^{a}	$E_{\rm loss}$	ΔE_1	$\Delta E_2^{\ b}$	$\Delta E_3 c$		
Active layer	(eV)	(eV)	(eV)	(eV)	(eV)	EQE _{EL}	$E_U(eV)$

PM6: BQx-O	1.461	0.549	0.264	0.043	0.245	7.63×10 ⁻⁴	22.25
PM6:BQx-Se	1.484	0.578	0.266	0.052	0.262	3.96×10-4	24.05
PM6: BQx-S	1.482	0.559	0.266	0.054	0.254	5.47×10-4	23.29

^{*a*} E_{g} : determined from the derivatives of the EQE curve and the mean peak energy. ^{*b*} ΔE_{2} : voltage losses due to non-ideal absorption (it was calculated from EL and FTPS measurements). ^{*c*} ΔE_{3} : voltage losses due to non-radiative recombination only.

Table S6. Charge mobility parameters of the optimized PM6: BQx-O, PM6: BQx-Se and PM6:BQx-S devices.

Device	μ_h (cm ² V ⁻¹ s ⁻¹)	μ_e (cm ² V ⁻¹ s ⁻¹)	μ_{h/μ_e}	$P_{\rm diss}(\%)$	$P_{\text{coll}}(\%)$
PM6: BQx-O	4.75 × 10 ⁻⁴	4.12×10^{-4}	1.15	99.1	90.9
PM6:BQx-Se	3.52×10^{-4}	2.61 × 10 ⁻⁴	1.35	94.4	69.1
PM6: BQx-S	4.21 × 10 ⁻⁴	3.32×10^{-4}	1.27	96.9	77.7

Table S7. Morphology parameters extracted from the GIWAXS measurements.

Film	q (010, Å ⁻¹)	d-space ^{<i>a</i>} (010, Å)	CCL ^b (010, Å)	q (100, Å-1)	d-space ^{<i>a</i>} (100, Å)	CCL ^b (100, Å)
BQx-O	1.71	3.67	25.47	0.34	18.37	188.50

BQx-Se	1.68	3.73	16.58	0.35	17.80	29.92	
BQx-S	1.69	3.72	18.60	0.34	18.48	74.41	
PM6: BQx-O	1.73	3.63	20.41	0.30	20.87	78.54	
PM6: BQx-Se	1.73	3.64	15.66	0.30	21.08	99.21	
PM6: BQx-S	1.74	3.61	19.17	0.30	21.01	89.76	

^{*a*} Calculated from the equation: d-spacing = $2\pi/q$. ^{*b*} Obtained from the Scherrer equation: CCL = $2\pi K/FWHM$, where FWHM is the full-width at half-maximum and K is a shape factor (K = 0.9 here).

 Table S8. The fitted parameters for PL decays of pure acceptor films.

Sample	\mathbf{A}_1	τ_1 , ps	A_2	τ_2 , ps
BQx-O	0.36	0.55	0.63	0.823
BQx-Se	0.45	0.483	0.55	0.803
BQx-S	0.54	0.442	0.45	0.859

Table S9. The fitted parameters for donor GSB of blended films.

Sample	A ₁	τ_1 , ps	A ₂	τ_2 , ps
PM6: BQx-O	0.23	0.93	0.76	7.95
PM6:BQx-Se	0.33	0.5	0.63	15.33
PM6: BQx-S	0.33	0.5	0.65	12.53

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