Electronic supplementary information for the manuscript:

"Environment-friendly aqueous processing of [60]fullerene semiconducting films for truly green organic electronics"

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Experimental

Materials and solvents

Fullerene C₆₀ (99.5%) was obtained from Fullerene Center Ltd., Nizhniy Novgorod, Russia. Iodine monochloride (98%), diisopropylethylamine (98%), 2-(Bocamino)ethanethiol (97%), 3-mercaptopropionic acid (99%), trifluoroacetic acid (99%), 1,2-dichlorobenzene (99%), were obtained from Acros Organics. The dielectric CYCLOTENE 4024 resin (BCB) was obtained from The Dow Chemical Company. Mesitylene, citric acid (puriss. spec.), toluene (puriss. spec.) were purchased from Chimmed.

Synthesis of F1 and F2 fullerene derivatives

Compounds F1 (F1a) and F2 (F2a) were synthesized according to the scheme S1 following the previously reported procedures. (E. A. Khakina, *et. al, Chem. Commun.*, 2012, 48, 7158.)



Scheme S1 Synthesis of fullerene derivatives F1 and F2.

Synthesis of F1

To obtain the derivative F1500 mg of compound F1a was stirred at room temperature in 30 ml of trifluoroacetic acid for 30 minutes. Then the solvent was removed under vacuum. The dark-red residue was washed with diethyl ether and dried in air. The product yield was 95%.

Synthesis of F2

Compound F2 was obtained by dissolving F2a (120 mg) in aqueous ammonia (30%, 10 ml). The excess of ammonia was removed under vacuum. The dark red aqueous solution of F2 was filtered through PES syringe filter (0.45 μ m, ISOLAB) and freeze-dried.

Fabrication of OFETs

Glass slides were used as substrates for fabrication of OFETs. Substrate cleaning, deposition of aluminum gate electrodes followed by Al anodization to form AlO_x dielectric layer and subsequent deposition of CYCLOTENE 4024 resin (BCB) as secondary dielectric were performed according to the procedures reported previously (L. I. Leshanskaya *et al.*, *Adv. Opt. Mater.*, 2017, **5**, 1601033).

Compounds F1 and F2 were dissolved in the mixture of deionized water and isopropanol (1:1 vol. ratio) in concentration of 20 mg/ml. The resulting solutions of F1 and F2 were filtered through PES syringe filters (0.45 μ m, ISOLAB). To form the semiconducting fullerene layer, a solution of the fullerene derivative (F1 or F2) was spin coated on top of the dielectric layer at 1000 rpm in air. The films were then transferred inside the glovebox and annealed for 30 minutes (annealing temperatures are specified in the main text).

Silver drain and source electrodes were thermally evaporated on top of the semiconductor fullerene film at the base pressure ~ 5×10^{-6} mbar. The deposition rate was kept at ~ 0.5-1 Å/s. The channel length and width were 75 µm and 2 mm, respectively.

Characterization techniques

Transfer and output characteristics of the fabricated OFETs were measured using Kethley 2612A instrument with LabTracer software.

UV-Vis absorption spectra were measured using an AvaSpec-2048-2 dual- channel fiber spectrometer.

Gel-permeation chromatography was performed using Phenogel 5u 100 A, 3.0×300 mm column (Phenomenex) and Shimadzu LC20 instrument. Toluene-acetonitrile 99:1 v/v mixture was used as the eluent with flow rate 0.3 mL/min

X-ray diffraction analysis was performed using Bruker D8 Advance X-Ray diffractometer with CuK_{α} radiation and LYNXEYE XY Detector in the 2 θ range 0-60°.

Scanning electron microscopy (SEM) images were obtained on a Zeiss SUPRA 25 instrument.

Atomic force microscopy (AFM) images were obtained using NTEGRA PRIMA NT-MDT microscope.



Fig. S1 Thermal gravimetry curve for compound F2



Fig. S2 UV-Vis absorption spectra of pristine and annealed films of (a) **F1** and (b) **F2.**



Fig. S3 Gel-permeation chromatogram of pristine **F2** (red), reference C_{60} (black) and **F2** annealed at 300°C (blue).



Fig. S4 The ¹³C NMR spectrum of C_{60} produced by thermal annealing of thin film of F1 (CS₂ - C_6D_{12} , 20 : 1 v/v ratio, 125 MHz).







Fig. S6 SEM images of F2 films annealed at different temperatures.



Fig. S7 AFM images of F1 films annealed at different temperatures.



Fig. S8 AFM images of F2 films annealed at different temperatures.