Electronic Supplementary Material (ESI) for RSC Advances.

Naphthalene Diimide-based *n*-type Small Molecule Organic Mixed Conductors for Accumulation Mode Organic Electrochemical Transistors

Seongdae Kang[1], Jiaxin Fan[2], João B.P. Soares[1], Manisha Gupta[2,]*

[1]Department of Chemical and Materials Engineering, University of Alberta, Edmonton, Alberta, T6G 1H9, Canada.

[2]Department of Electrical and Computer Engineering, University of Alberta, Edmonton, Alberta, T6G 1H9, Canada.

[*] Corresponding author email address: mgupta1@ualberta.ca

Supporting Information

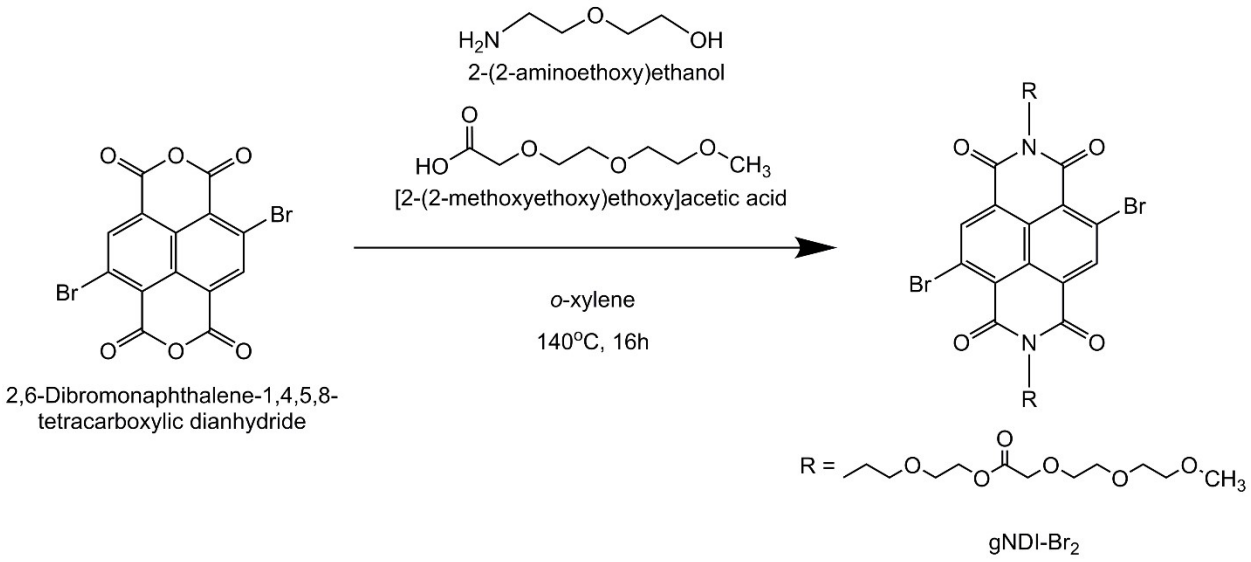


Figure S1. A synthesis scheme of the gNDI-Br_2.

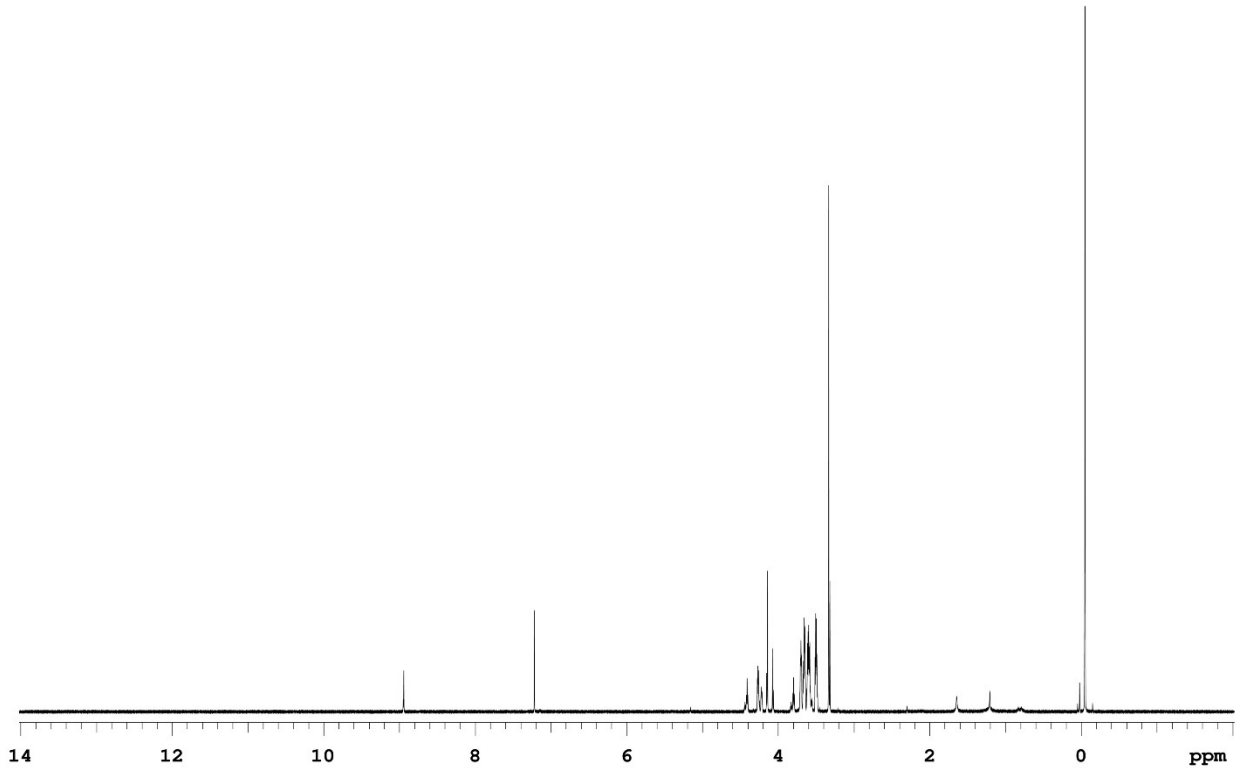


Figure S2. ^{1}H-NMR spectrum of the gNDI-Br_2 in $CDCl_3$.

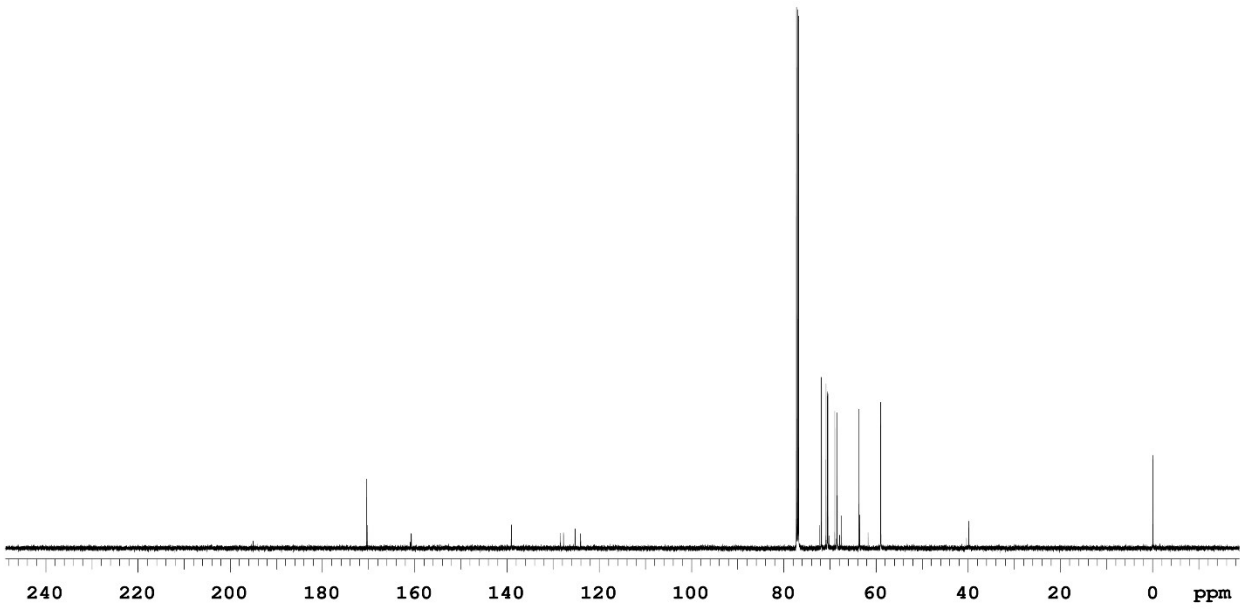


Figure S3. ^{13}C-NMR spectrum of the gNDI-Br_2 in $CDCl_3$.

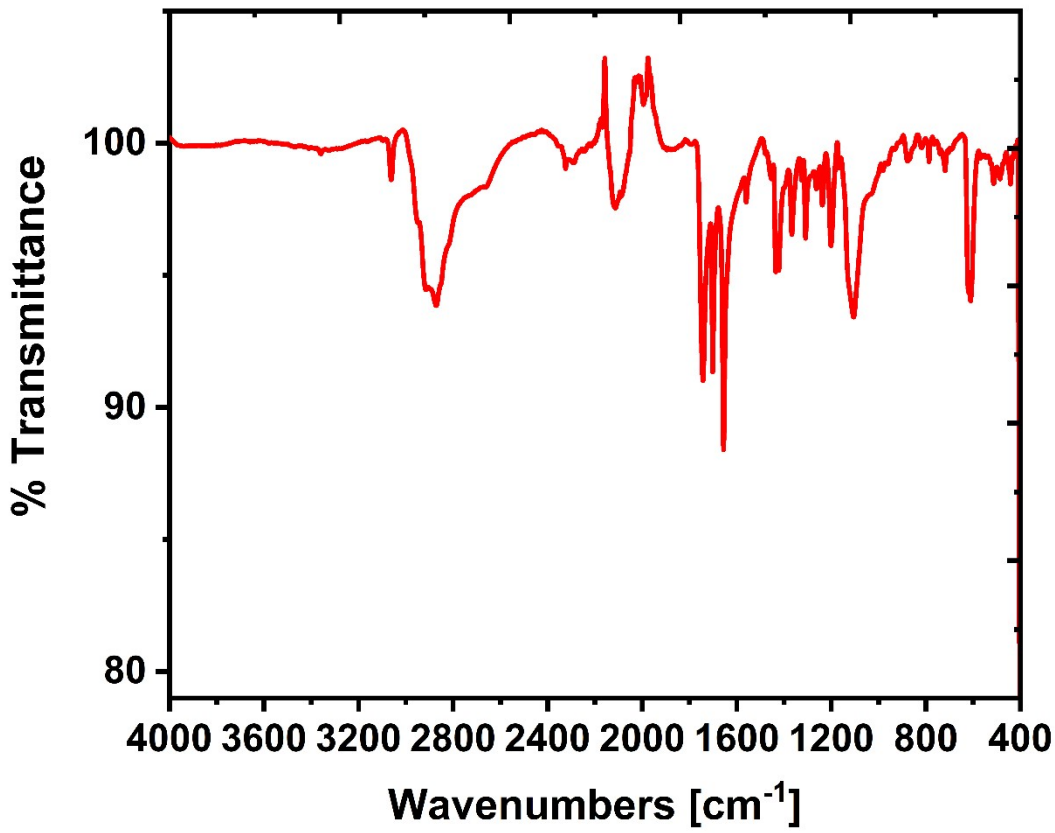
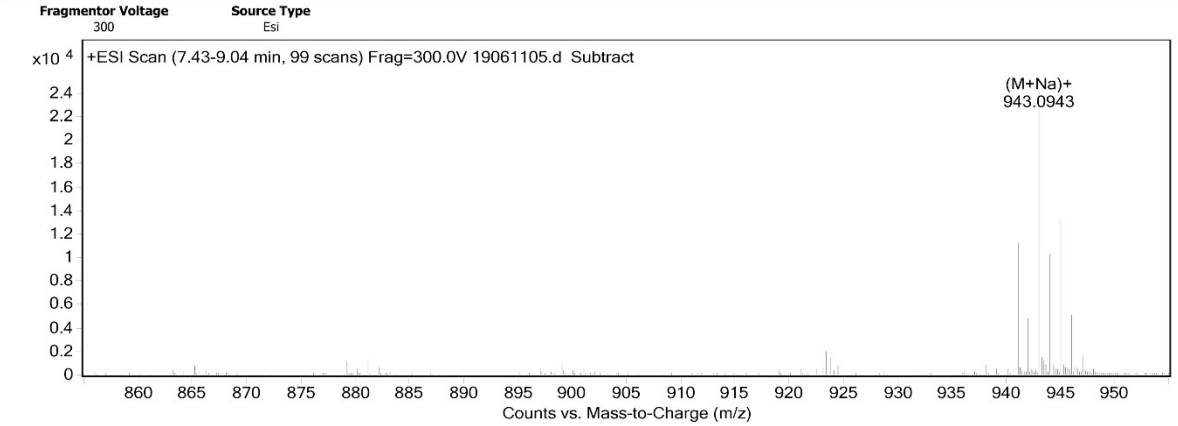


Figure S4. Attenuated total reflection Fourier Transform Infrared spectroscopy spectra of the gNDI-Br_2.

User Spectra



Formula Calculator Results

Formula	Ion Species	Mass	Calc. Mass	m/z	Calc. m/z	Diff (mDa)	Diff (ppm)	DBE	Ion	Score
C36 H44 Br2 N2 O16	C36 H44 Br2 N2 Na O16	918.1067	918.1058	941.0958	941.095	-0.82	-0.87	15	(M+Na)+	98.63

Figure S5. Electrospray Ionization Time-of-Flight Mass spectroscopy spectra of the gNDI-Br_2.

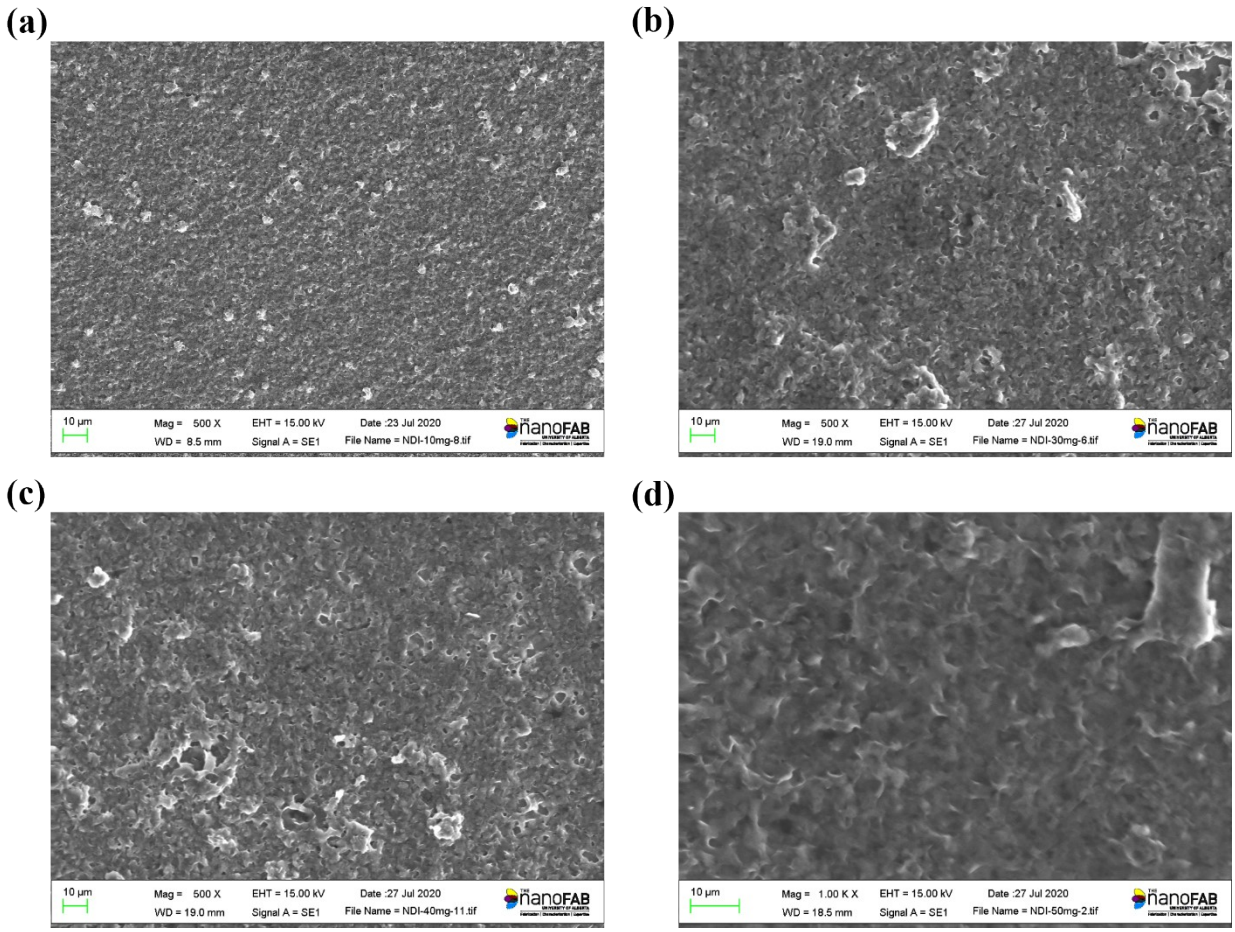


Figure S6. Scanning electron microscopy (SEM) images of the thin-film gNDI-Br_2 with different concentrations. a) 10 mg mL^{-1}, b) 30 mg mL^{-1}, c) 40 mg mL^{-1}, d) magnified (1000x) image of 50 mg mL^{-1}.

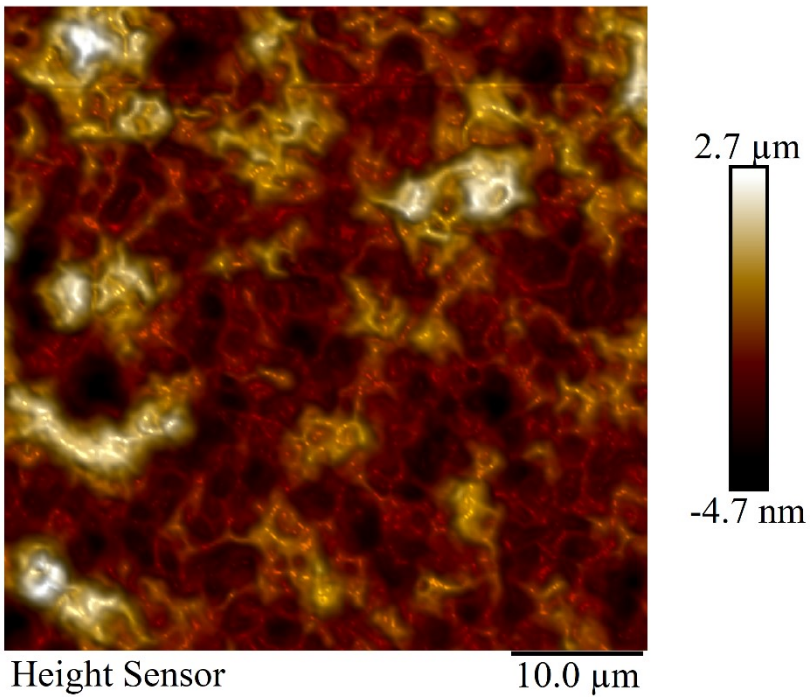


Figure S7. Roughness measurement of the drop-casted gNDI-Br_2 (50 mg mL^{-1}) thin film by atomic force microscopy (AFM) and root-mean-square roughness (R_q)of the thin film is 375 nm.

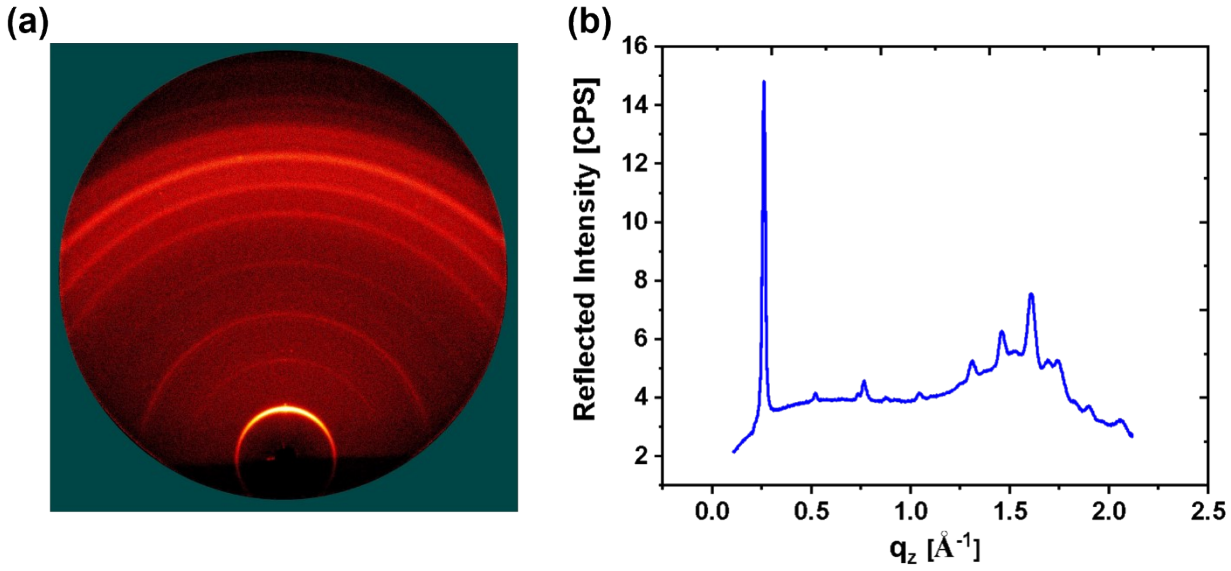


Figure S8. (a) Grazing angle Incidence X-ray diffraction pattern of the gNDI-Br_2, (b) GIXRD line cuts along with q_z direction.

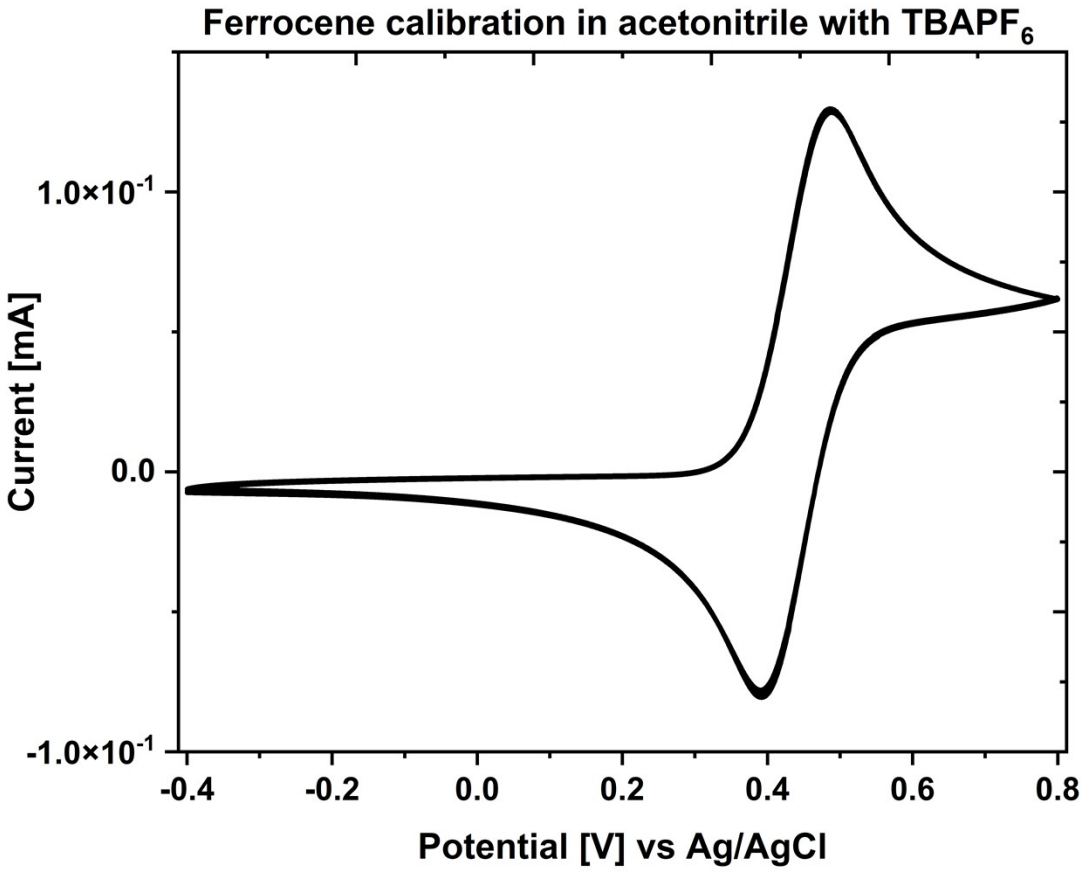


Figure S9. The cyclic voltammogram for $K_4Fe(CN)_6$ in acetonitrile with 100 mM of tetrabutylammonium hexafluorophosphate ($TBAPF_6$) supporting electrolytes.

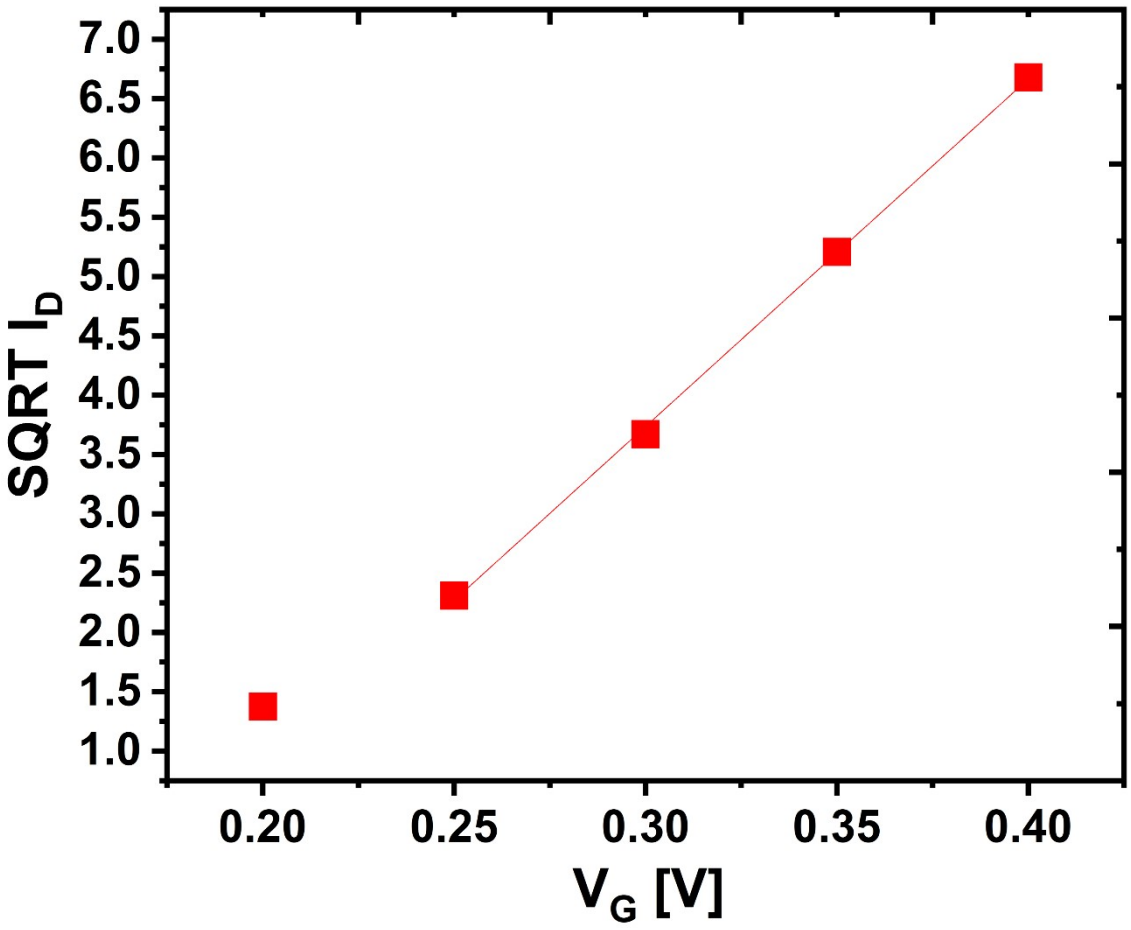


Figure S10. The threshold voltage of the device using sqrt(I_D) vs V_G. Threshold voltage, V_T = 172.5 mV.

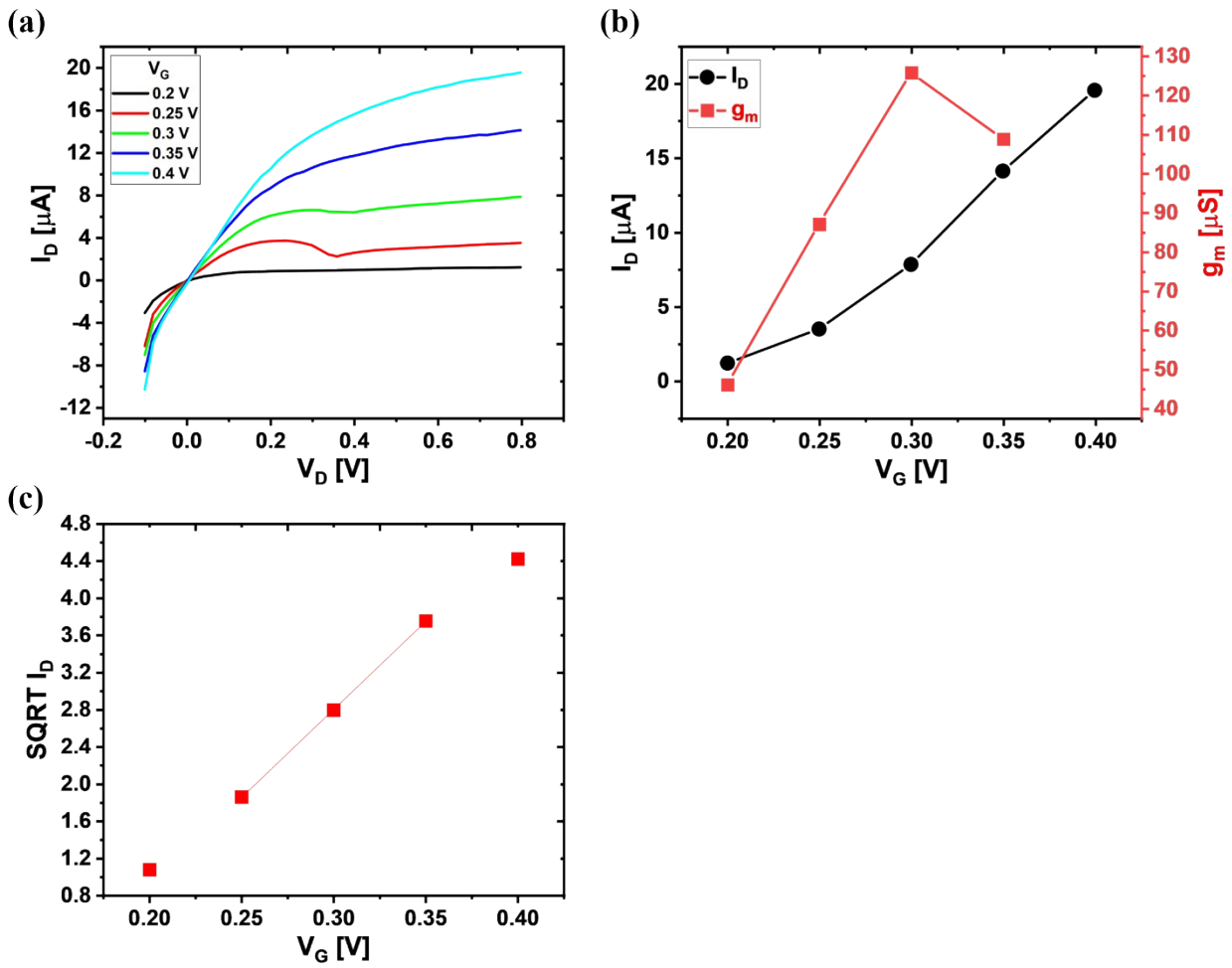


Figure S11. (a) Output characteristics of the n-type gNDI-Br_2 (50 mg mL^{-1}) OECT measured with 100 mM NaCl electrolyte and Ag/AgCl gate electrode with the channel width (W) of 871 µm, length (L) of 20 µm, and thickness (d) of 1.3 µm. (b) Transfer curve and transconductance of the same OECT device. (c) The threshold voltage of the device using sqrt(I_D) vs V_G. Threshold voltage, V_T = 151.9 mV.

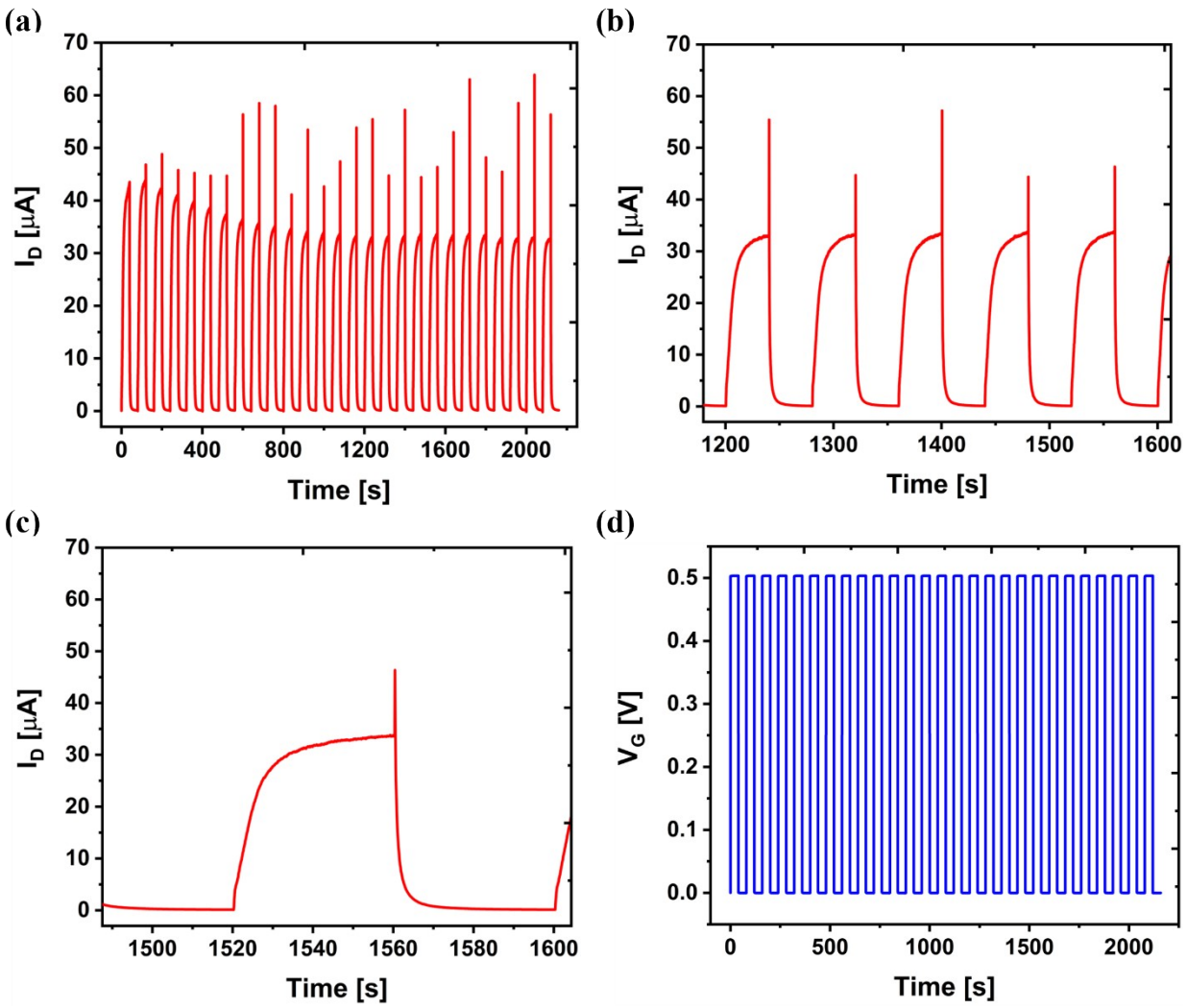


Figure S12. The repeated cycling stability measurement with the gNDI-Br_2 (50 mg mL^{-1}) OECT device. (a) the complete cycle of I_D vs time during 36 min. (b) magnified I_D vs time for 7 min. (c) a single pulse of the long-term stability measurement. (d) V_G vs time.

Supplementary methods

Materials. 2,6-dibromonaphthalene-1,4,5,8-tetracarboxylic dianhydride (NDA-Br_2) was purchased from TCI America. 2-(2-aminoethoxy)ethanol, 2-(2-methoxyethoxy)ethoxy acetic acid,

anhydrous *o*-xylene and chloroform were purchased from Sigma-Aldrich. Deuterated chloroform ($CDCl_3$) was purchased from Sigma-Aldrich.

Synthesis. NDA-Br_2 (525 mg, 1.78 mmol) was charged into the thoroughly dried 50 mL two-neck round bottom flask. The flask was connected with the Schlenk line to make the inert atmosphere under nitrogen. 20 mL of *o*-xylene was added into the flask, then 2-(2-aminoethoxy)ethanol (0.4 mL, 4.0 mmol) and 2-(2-methoxyethoxy)ethoxy acetic acid (3.5 mL, 33 mmol) was added dropwise into the stirred solution. The reaction mixture was heated to 140°C for 16 hours. The reactant was cooled to room temperature and concentrated under reduced pressure. 50 mL of $CHCl_3$ was added and washed with 100 mM HCl, 100 mM $NaHCO_3$, brine and DI water sequentially. The organic solution was dried with $MgSO_4$, and the solvent was removed *in vacuo*. The reactant was purified with column chromatography with ethyl acetate:methanol (98:2). The amorphous yellow solid was extracted from the removing solvent *in vacuo*. 139 g of the material was acquired with a yield of 26 %. HRMS (ESI-ToF): 918.1067 ($M+Na^+$) with the calculated mass of 918.1058. ^{1}H-NMR (600 MHz, $CDCl_3$): 8.94 (s, 2H), 4.42 – 4.4 (t, J = 5.6 Hz, 4H), 4.28 – 4.25 (m, 4H), 4.14 (s, 4H), 4.08 (s, 4H), 3.81 – 3.79 (t, J = 6.0 Hz, 4H), 3.71 – 3.69 (m, 4H), 3.67 – 3.64 (m, 4H), 3.63 – 3.58 (m, 4H), 3.52 – 3.49 (m, 4H), 3.34 (s, 6H) ppm. ^{13}C-NMR (150 MHz, $CDCl_3$): 170.3, 160.8, 160.7, 139, 128.4, 125.2, 124, 71.8, 70.8, 70.5, 70.4, 68.9, 68.4, 67.5, 63.6, 58.9, 39.9 ppm. ATR FT-IR: significant peaks at 3062, 2871, 1743, 1702, 1656, 1435, 1422, 1367, 1309, 1202, 1106, 611 cm^{-1}.