Electronic Supporting Information

Electrostatically Driven Unidirectional Molecular Flux for High Performance Alkaline Flow Batteries

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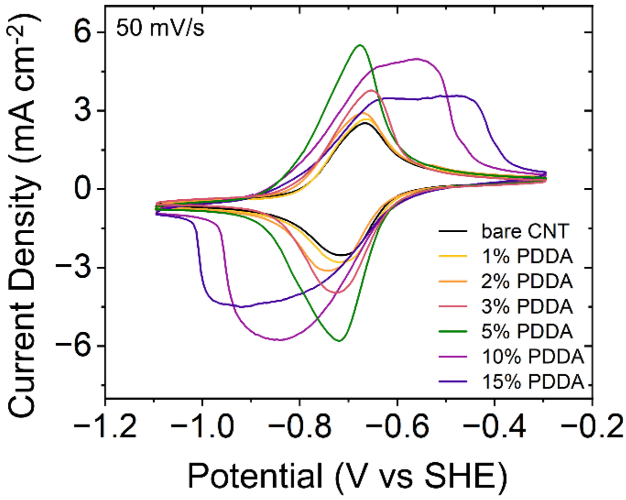


Fig. S1 Cyclic voltammogram of 2,6-DHAQ molecule on the CNT (+) electrode with varying concentrations of PDDA in the modifying solution.

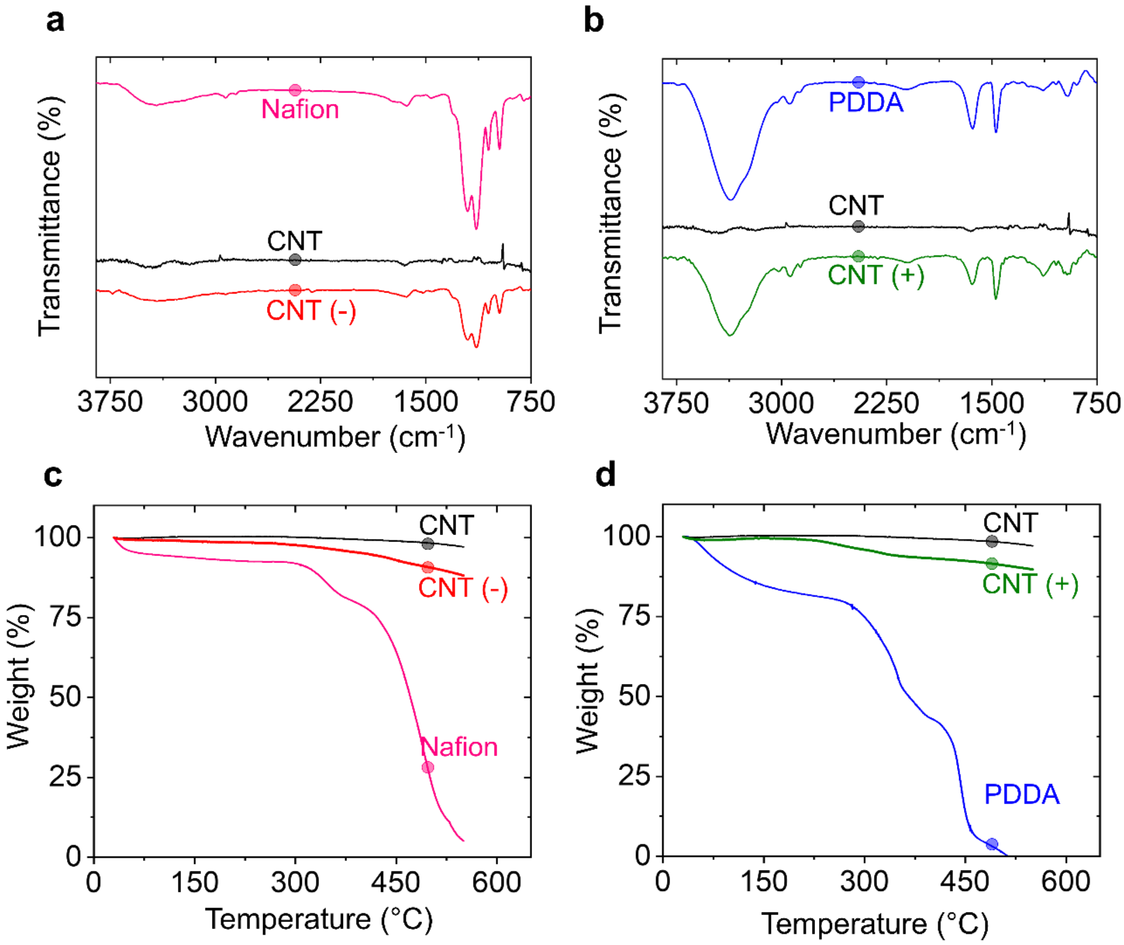


Fig. S2 (a) ATR-FTIR spectra of CNT (-) and individual components. **(b)** ATR-FTIR spectra of CNT (+) and individual components. **(c)** TGA of CNT (-) and individual components. **(d)** TGA spectra of CNT (+) and individual components.

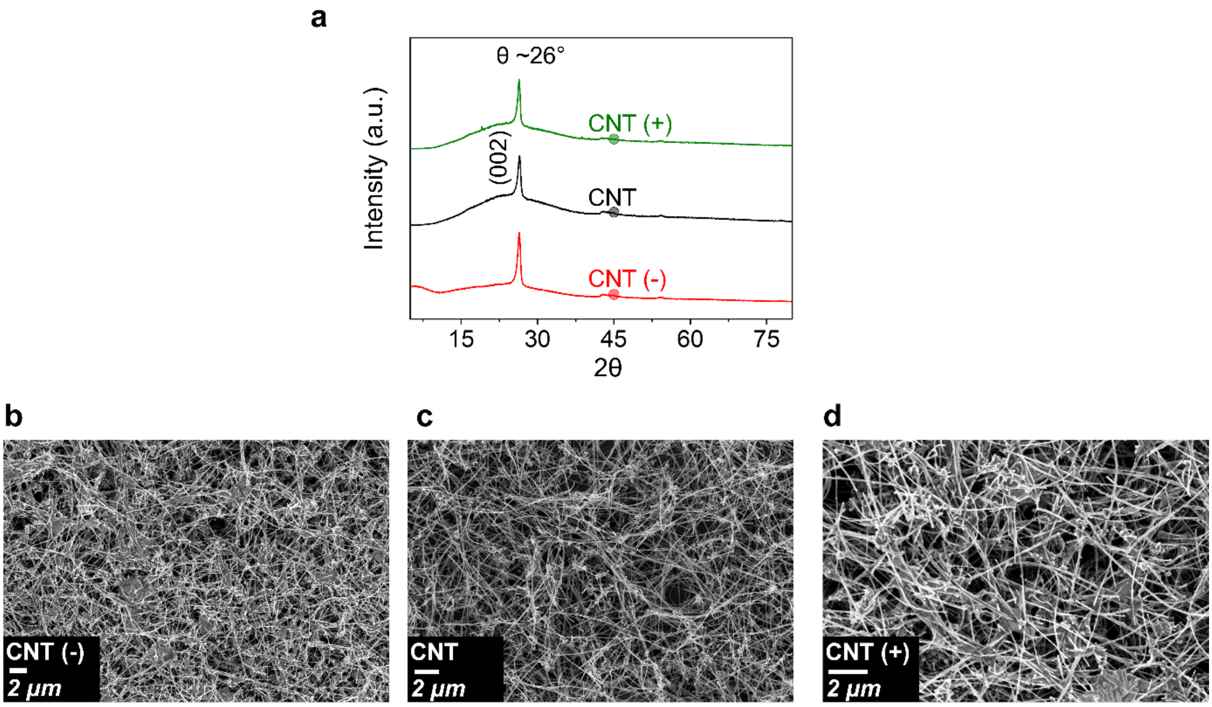


Fig. S3 (a) Powder-XRD for CNT (+), CNT, and CNT (-). SEM images for **(b)** CNT (-), **(c)** CNT, and **(d)** CNT (+)

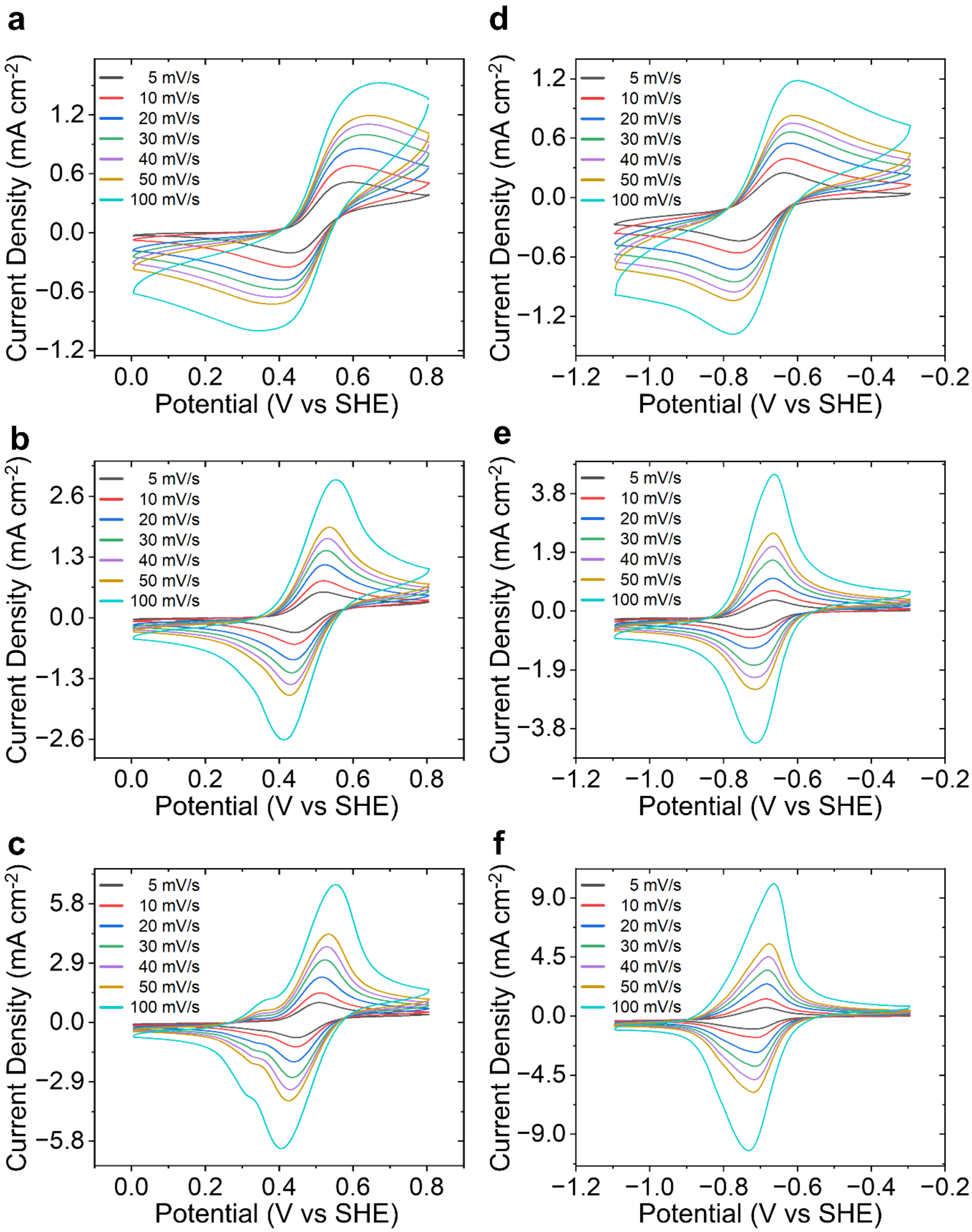


Fig. S4 Scan rate dependence studies in 10 mM $K_4[Fe(CN)_6]$ in 1M KOH with **(a)** CNT(-), **(b)** CNT, and **(c)** CNT(+). Scan rate dependence studies in 5 mM 2,6-DHAQ in 1 M KOH with **(d)** CNT(-), **(e)** CNT, and **(f)** CNT(+).

Calculation S1

Mass Transfer Coefficient (k_m) Calculation

$k_m = \frac{i}{nFAC}$

k_m = mass transfer coefficient (cm/s), i = limiting current (mA), n = number of electrons, A = area of electrode (cm^2), C = concentration (mol/cm^3), F = Faraday constant (~96485 C/mol),

(a) From Fig. 3b

For 10 mM $K_4Fe(CN)_6$ solution in 1 M KOH

C = 0.01 mmol/ cm^3

A = 0.196 cm^2

n = 1

- For CNT (-)

$k_m = \frac{i}{nFAC}$

$= \frac{4.667}{96485*0.01}$ = 0.00483 cm/s

- (ii) For CNT

$k_m = \frac{i}{nFAC}$

$= \frac{7.137}{96485*0.01}$ = 0.00739 cm/s

- (iii) For CNT (+)

$k_m = \frac{i}{nFAC}$

$= \frac{10.9}{96485*0.01}$ = 0.0113 cm/s

(b) From Fig. 3d

For 5 mM 2,6-DHAQ solution in 1 M KOH

C = 0.005 mmol/ cm^3

A = 0.196 cm^2

n = 2

- For CNT (-)

$$k_m = \frac{i}{nFAC}$$

$$= \frac{2.961}{2*96485*0.005} = 0.003 \text{ cm/s}$$

- (ii) For CNT

$$k_m = \frac{i}{nFAC}$$

$$= \frac{5.069}{2*96485*0.005} = 0.005 \text{ cm/s}$$

- (iii) For CNT (+)

$$k_m = \frac{i}{nFAC}$$

$$= \frac{7.388}{2*96485*0.005} = 0.0076 \text{ cm/s}$$

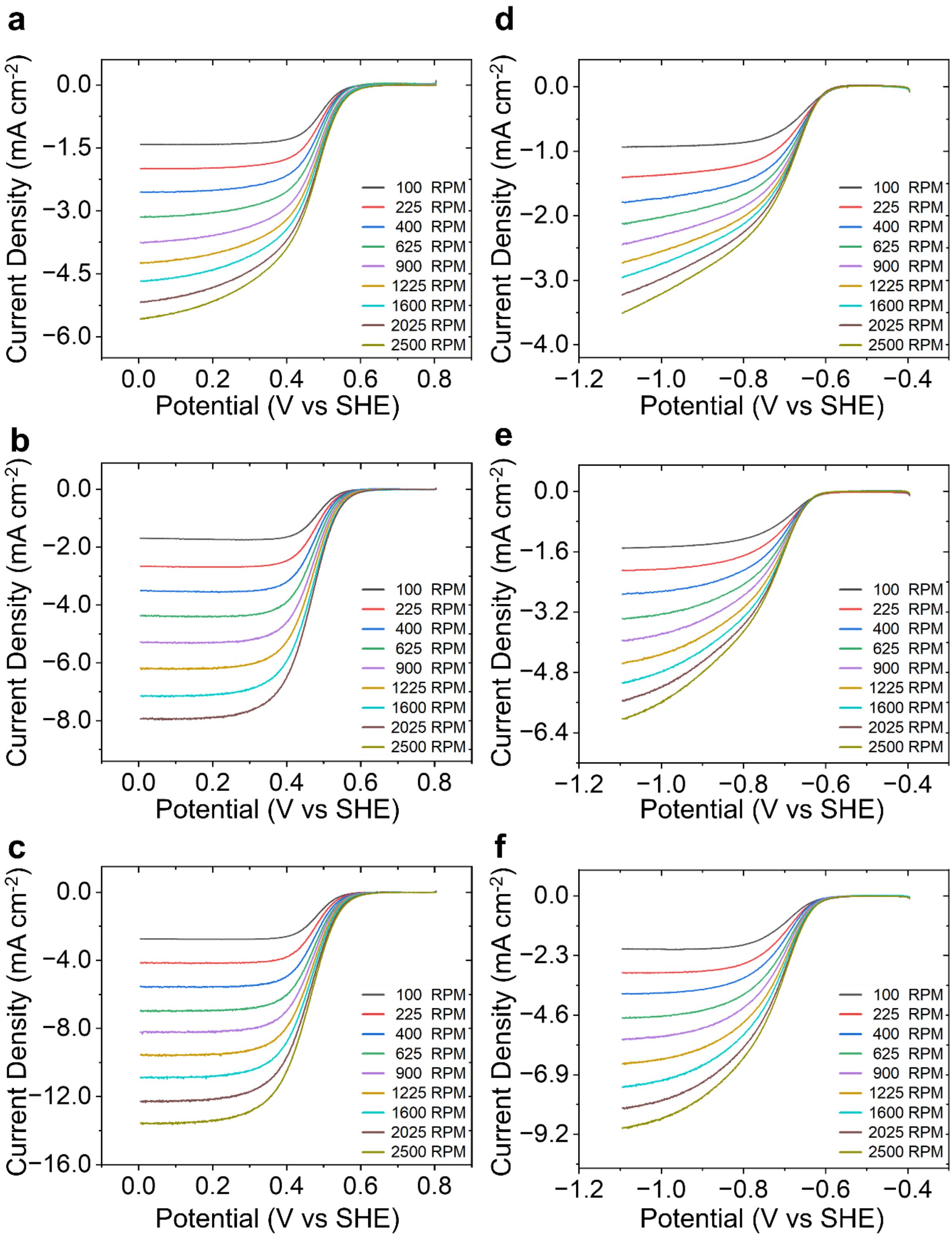


Fig. S5 RDE studies at different rotations for 10 mM $K_3[Fe(CN)_6]$ in 1 M KOH with **(a)** CNT (-), **(b)** CNT and **(c)** CNT (+). RDE studies at different rotations for 5 mM 2,6-DHAQ in 1 M KOH with **(d)** CNT (-), **(e)** CNT and **(f)** CNT (+).

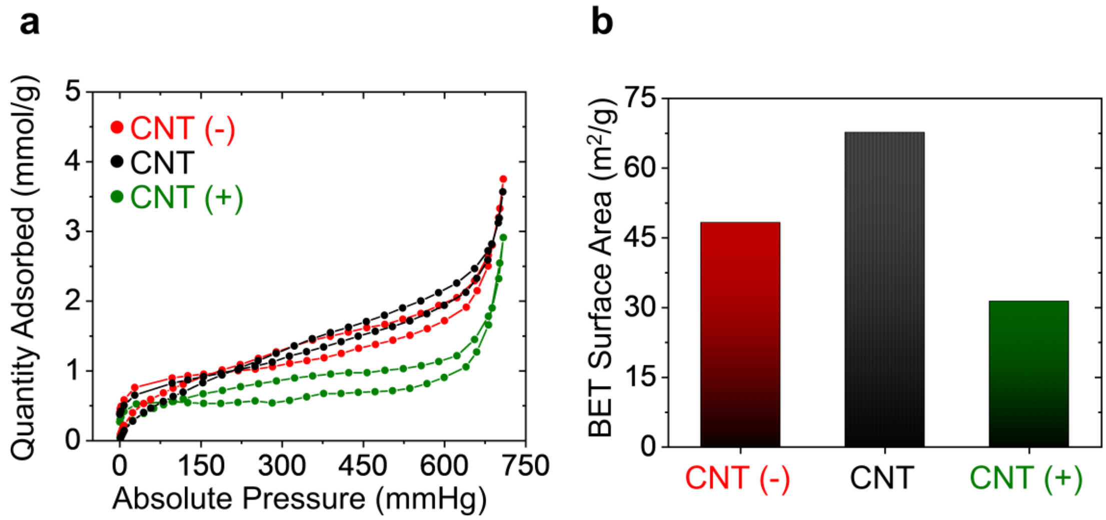


Fig. S6 (a) and **(b)** BET surface area for CNT (+), CNT and CNT (-).

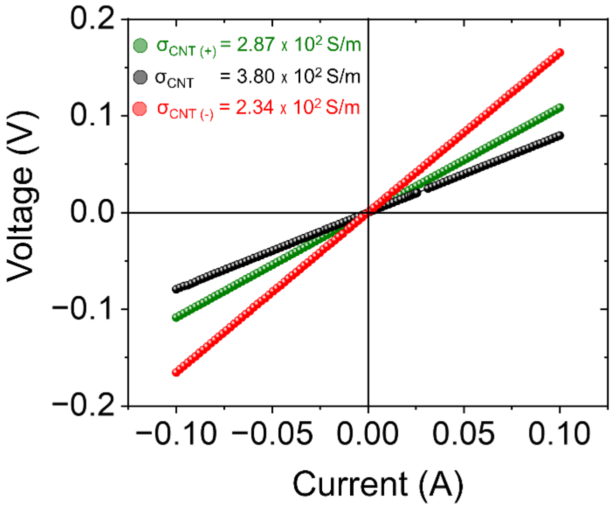


Fig. S7 I-V data for electrical conductivity measurements extracted from the four-point probe method for CNT (+), unmodified CNT and CNT (-).

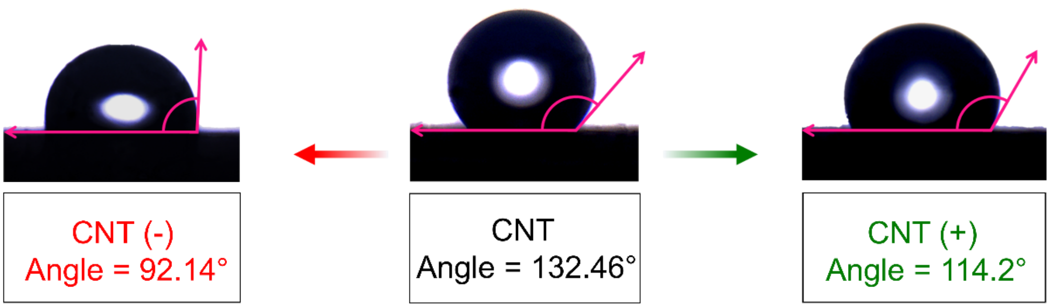
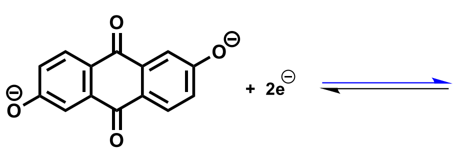


Fig. S8 Contact angle measurements for CNT (-), CNT and CNT (+) (Left to right).

The battery's half-cell and full-cell chemistries

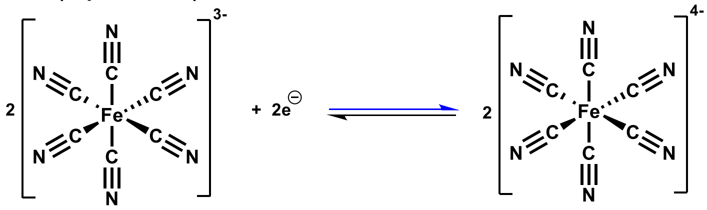
Anodic Reaction (Equation S1):

E° = -0.68 V vs SHE (pH 14)



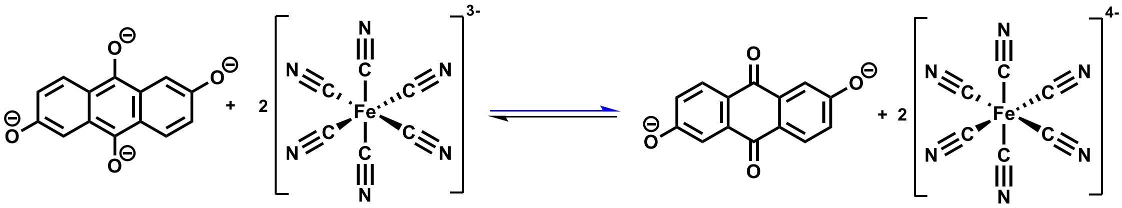
Cathodic Reaction (Equation S2):

E° = 0.36 V vs SHE



Overall Reaction (Equation S3):

$E°_{cell}$ = 1.04 V vs SHE



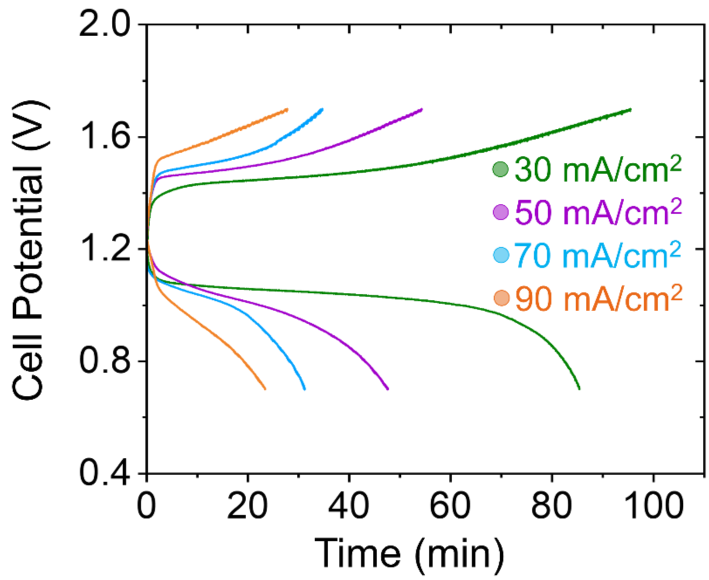


Fig. S9 Charge-discharge cycling of the redox flow battery equipped with the CNT (+) electrode at various current densities.

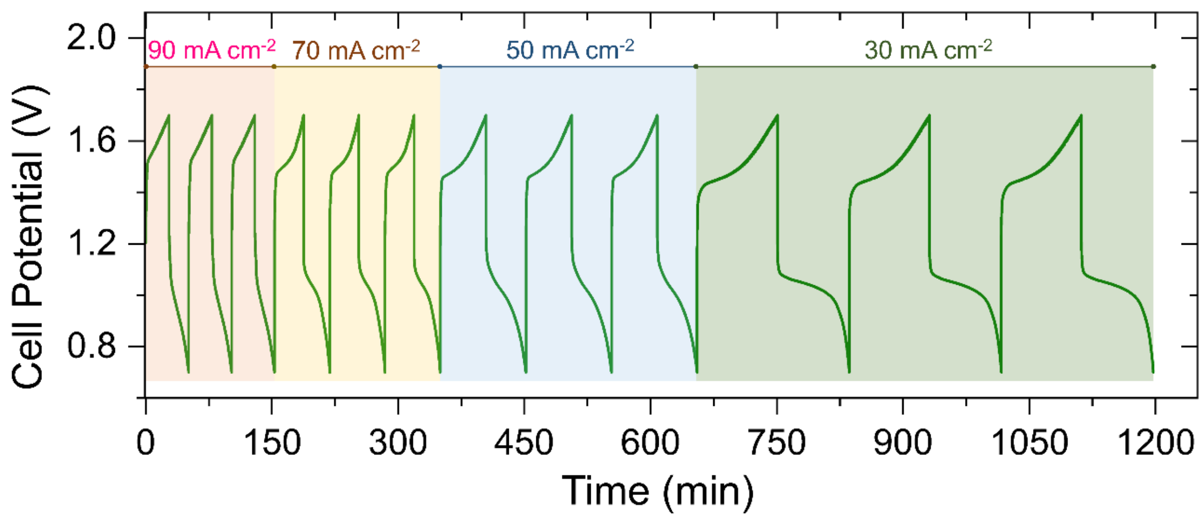


Fig. S10 Charge-discharge cycling of the redox flow battery equipped with the CNT (+) electrode at various current densities. Here, the data is shown as a function of time.

Calculation S2

Energy Efficiency (EE) Calculation

EE = $\frac{(C*V)_{discharge}}{(C*V)_{charge}}$

From Fig. 4d

- In case of CNT (-)

EE = $\frac{Area\ under\ the\ discharge\ curve\ (red\ discharge\ trace\ in\ Figure\ 4d)}{Area\ under\ the\ charge\ curve\ (red\ charge\ trace\ in\ Figure\ 4d)}$

= $\frac{208.89}{456.10}$

= 45.79 %

- In case of CNT

EE = $\frac{Area\ under\ the\ discharge\ curve\ (black\ discharge\ trace\ in\ Figure\ 4d)}{Area\ under\ the\ charge\ curve\ (black\ charge\ trace\ in\ Figure\ 4d)}$

= $\frac{465.36}{684.21}$

= 68.01 %

- In case of CNT (+)

EE = $\frac{Area\ under\ the\ discharge\ curve\ (olive\ green\ discharge\ trace\ in\ Figure\ 4d)}{Area\ under\ the\ charge\ curve\ (olive\ green\ charge\ trace\ in\ Figure\ 4d)}$

= $\frac{857.55}{933.23}$

= 91.89 %

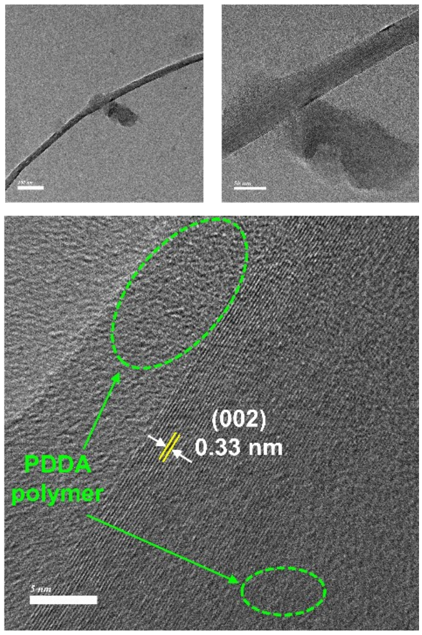


Fig. S11 TEM images of CNT (+) after charge-discharge cycling.

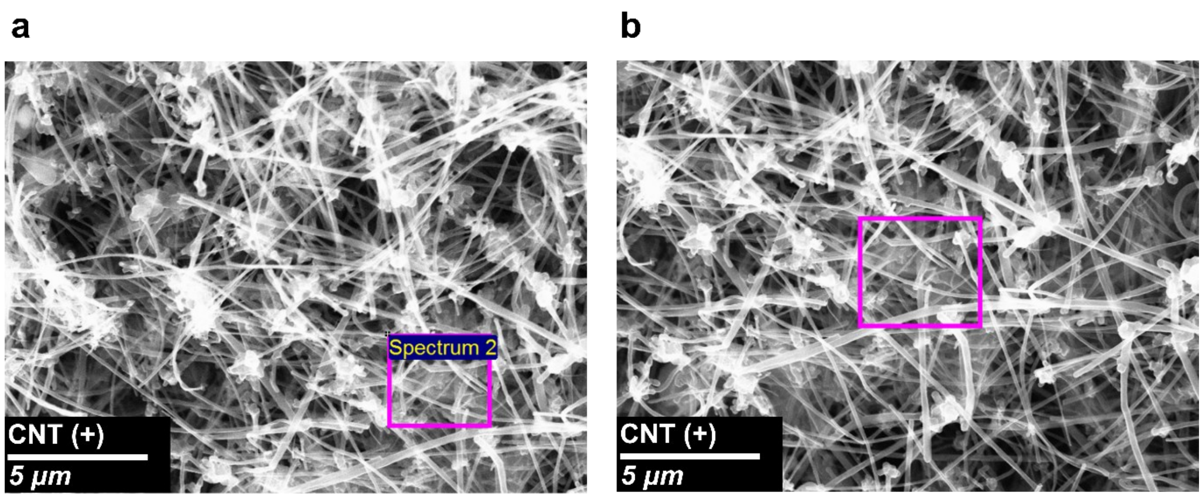


Fig. S12 SEM images of CNT (+) electrode utilized **(a)** at the cathodic interface and **(b)** at the anodic interface after long term charge-discharge.

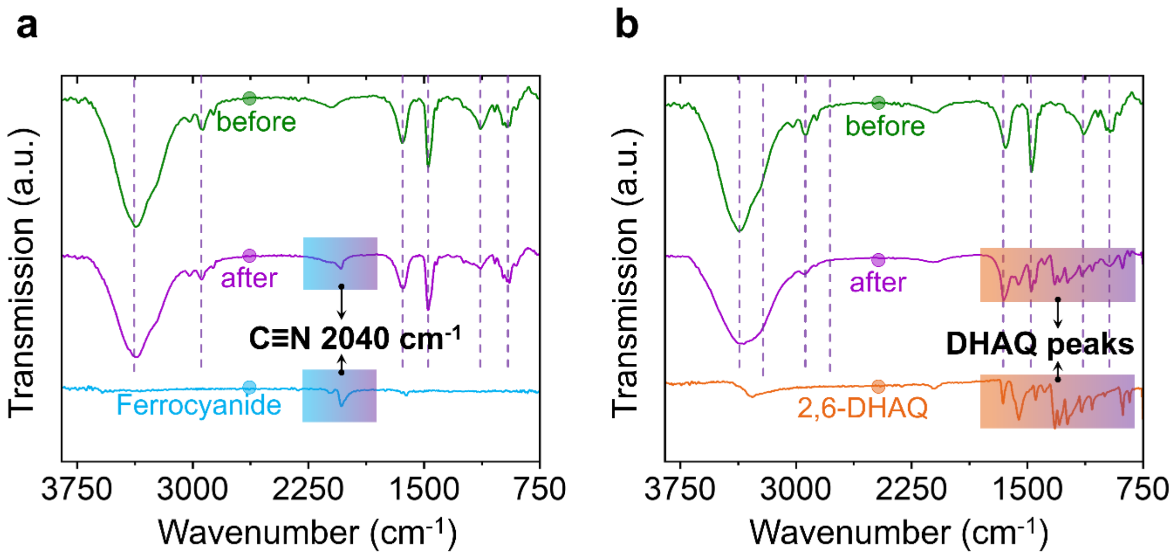


Fig. S13 ATR-FTIR spectra of CNT (+) electrode utilized **(a)** at the cathodic interface and **(b)** at the anodic interface before and after charge-discharge cycling.

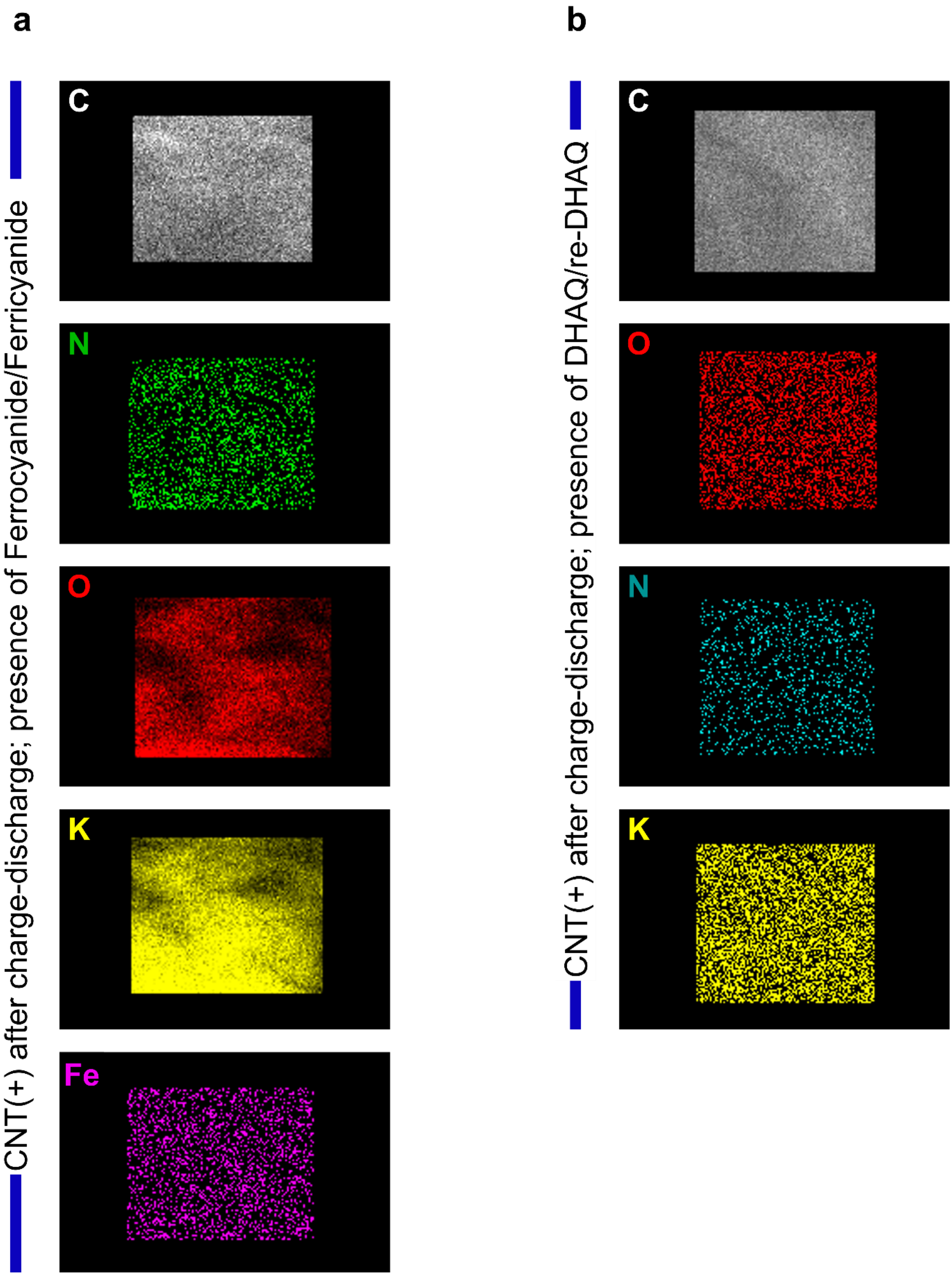


Fig. S14 EDX elemental mapping of CNT (+) electrode utilized **(a)** at the cathodic interface and **(b)** at the anodic interface after charge-discharge cycling. The signals of O, K and Fe in Fig. S14a originates from adsorbed iron species whereas the signals of K and O in Fig. S14b originates from adsorbed quinone molecule