

## Supplementary Materials

### Interfacial Co-polymerization Derived Nitrogen-doped Carbon Enables High-performance Carbon Felts for Vanadium Flow Batteries

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Table S1. Content of different nitrogen types on P-CF, DA-CF and PEI-DA-CF.

	pyridinic N (at.%)	pyrrolic N (at.%)	graphitic N (at.%)
P-CF	0.13	0.07	1.01
DA-CF	0.90	0.56	2.02
PEI-DA-CF	1.71	0.63	2.64

Table S2. Ohmic resistance ( $R_O$ ), charge transfer resistance ( $R_{CT}$ ) and diffusion resistance ( $R_D$ ) obtained by fitting Nyquist plots

	$R_O$ (mΩ)	$R_{CT}$ (mΩ)	$R_D$ (mΩ)
P-CF	71.81	117.4	22.86
DA-CF	71.38	77.75	21.68
PEI-DA-CF	74.2	65.26	12.8

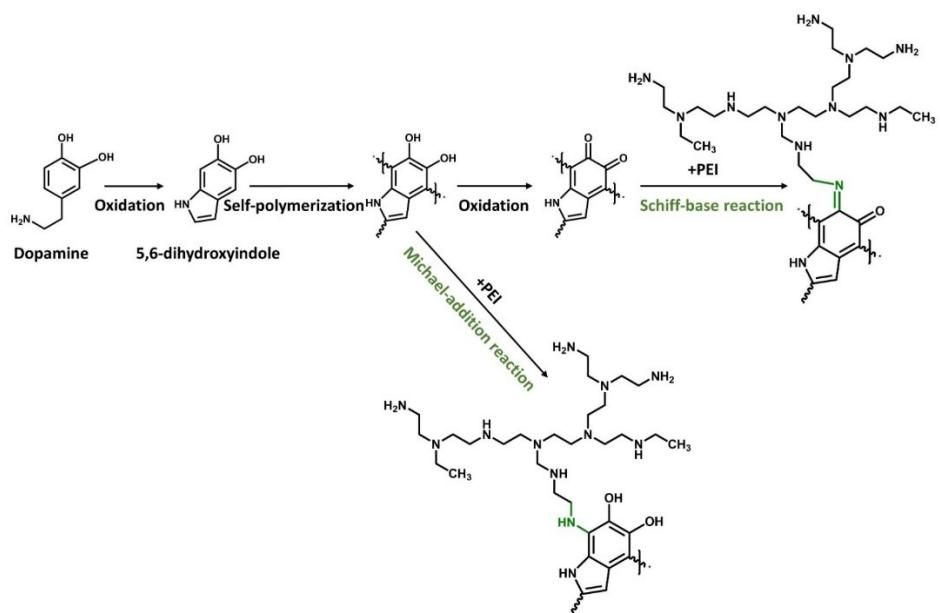


Figure S1. Proposed mechanism for the reactions between DA and PEI.

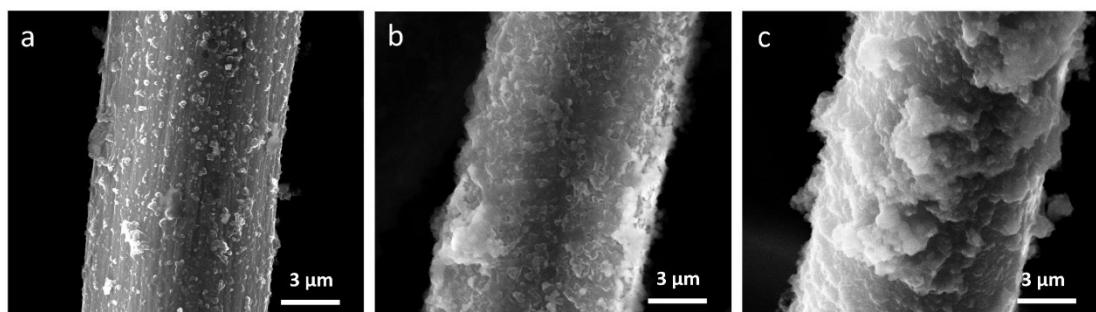


Figure S2. SEM images of Nitrogen-enriched precursor CFs with different co-polymerization time: (a) 10 h; (b) 30 h and (c) 50 h.

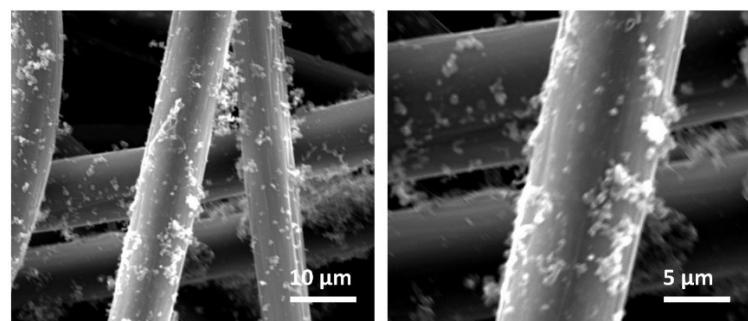


Figure S3. SEM images of Nitrogen-enriched precursor CF prepared via 30 h DA self-polymerization.

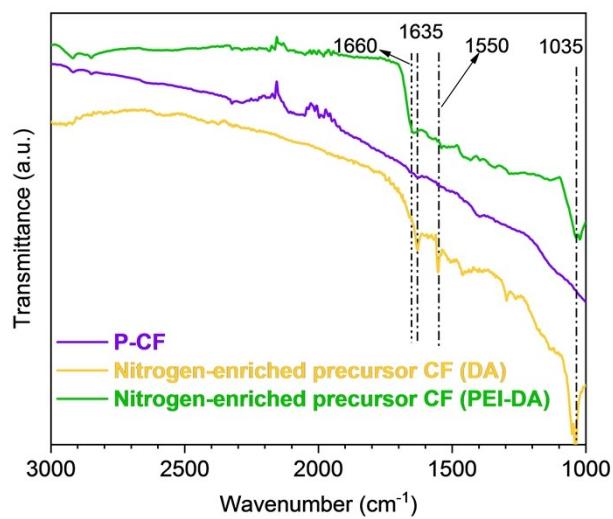


Figure S4. FT-IR spectra of P-CF and Nitrogen-enriched precursor CFs.

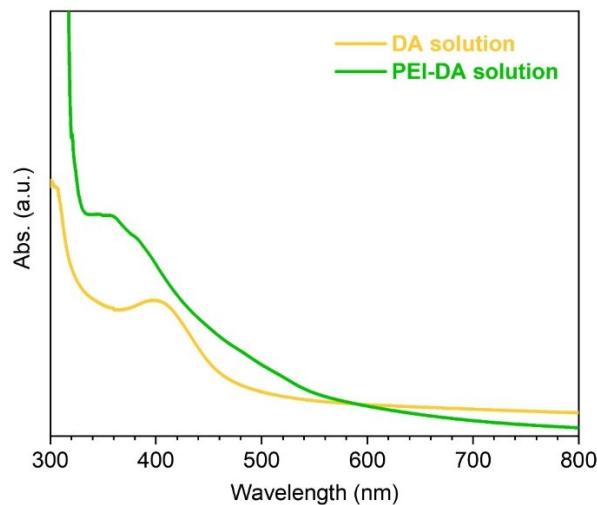


Figure S5. UV-Vis spectra of pure DA solution and PEI/DA solution.

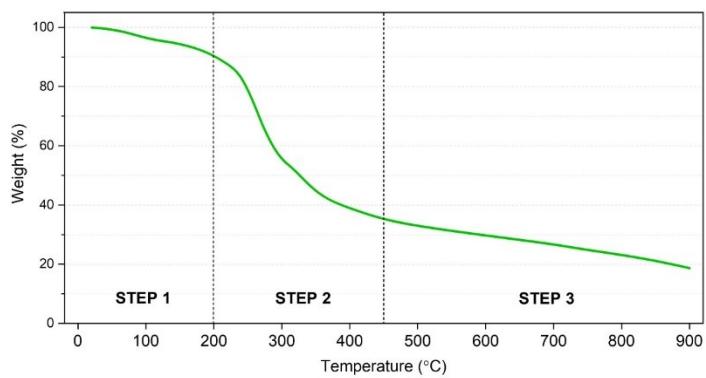


Figure S6. TGA curve of Nitrogen-enriched precursor layer.

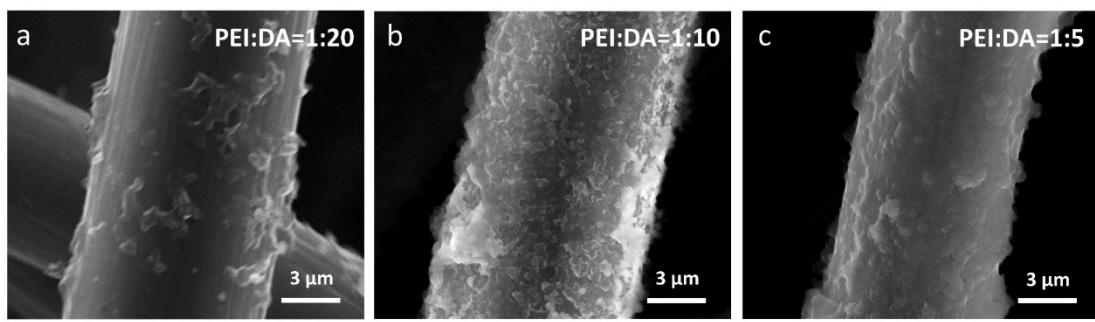


Figure S7. SEM images of Nitrogen-enriched precursor CFs with different mass ratios of PEI to DA: (a) 1:20; (b) 1:10 and (c) 1:5.

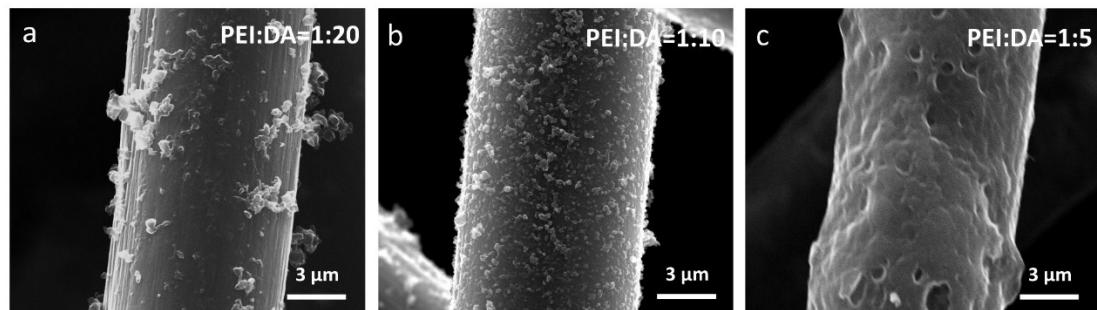


Figure S8. SEM images of PEI-DA-CF electrodes with different mass ratios of PEI to DA: (a) 1:20; (b) 1:10 and (c) 1:5.

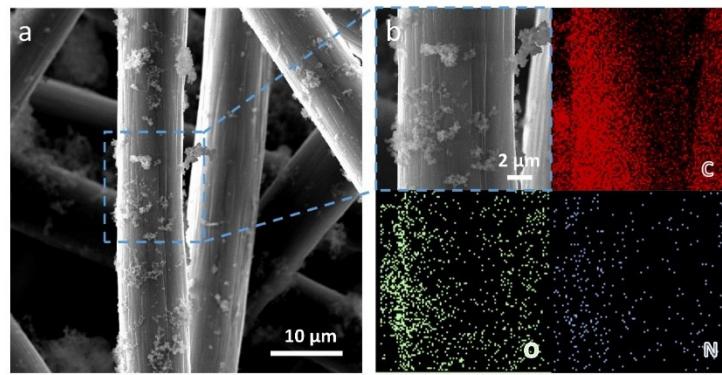


Figure S9. (a) SEM image of DA-CF and (b) corresponding EDX elemental mapping.

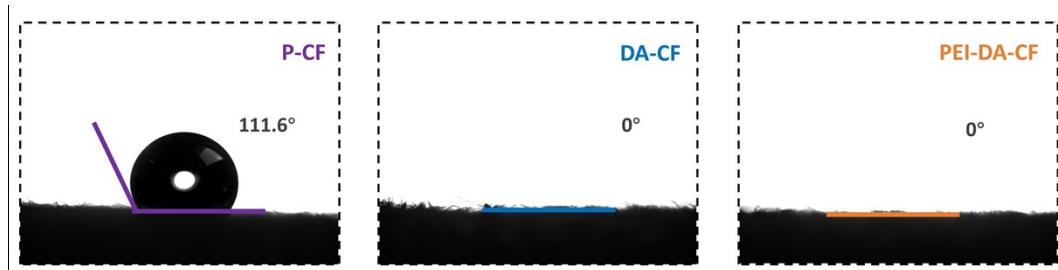


Figure S10. Contact angle measurements of P-CF, DA-CF and PEI-DA-CF.

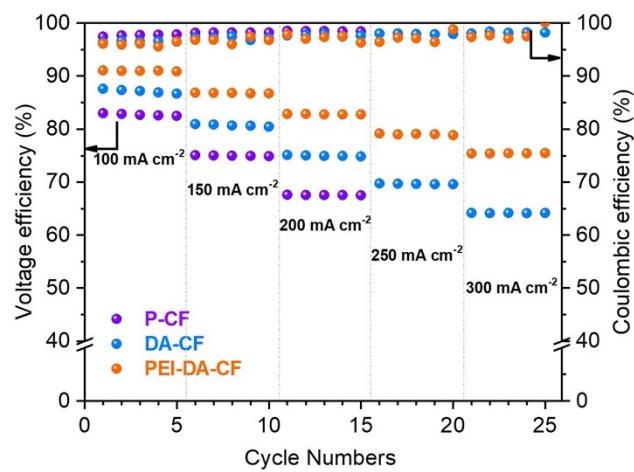


Figure S11. CE and VE of the VFBs at different current densities.

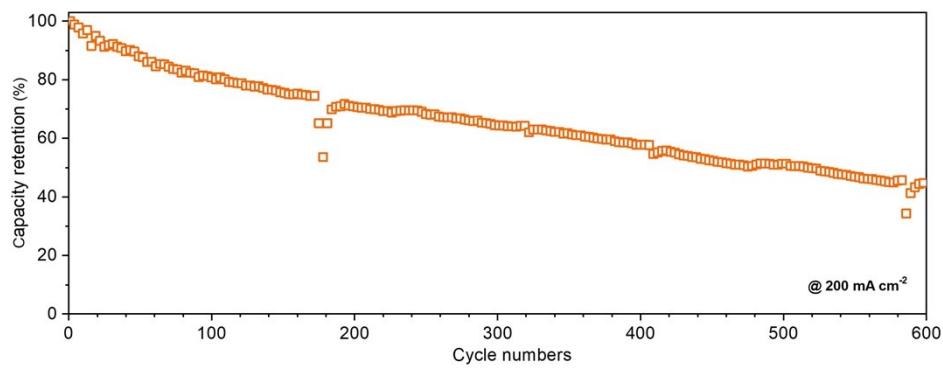


Figure S12. Capacity retention of the VFB with PEI-DA-CF electrodes at  $200 \text{ mA cm}^{-2}$ .

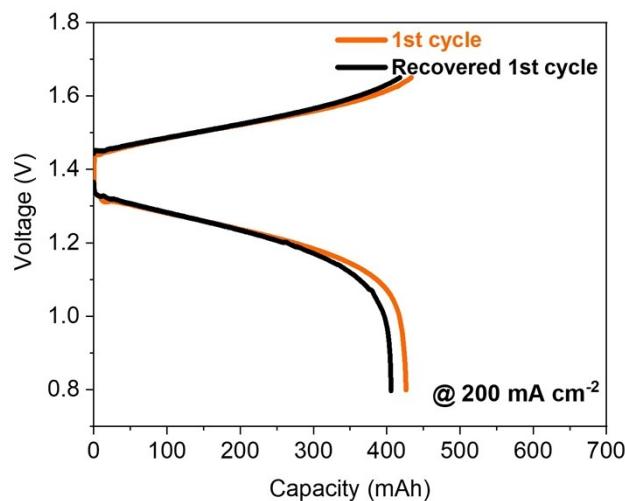


Figure S13. Comparison of charge-discharge curves for VFB with PEI-DA-CF electrodes before and after refreshing electrolytes and membrane at  $200 \text{ mA cm}^{-2}$ .

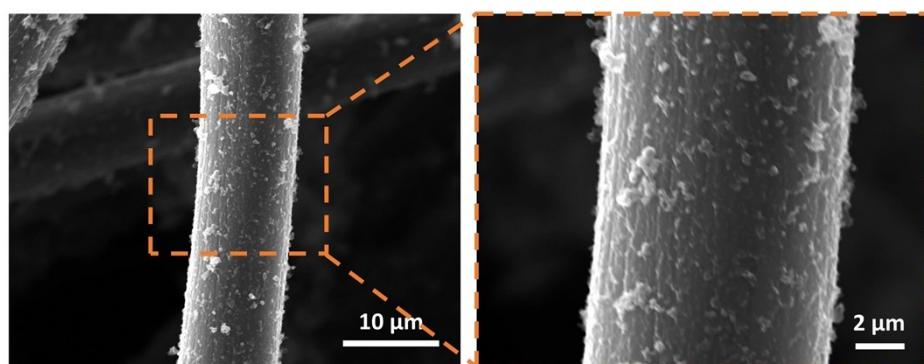


Figure S14. SEM images of PEI-DA-CF electrode after cycling at different magnifications.