

# ADVANCED FUNCTIONAL MATERIALS

## Supporting Information

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**Mass Transfer and Reaction Kinetic Enhanced Electrode  
for High-Performance Aqueous Flow Batteries**

*Alolika Mukhopadhyay, Yang Yang, Yifan Li, Yong Chen,  
Hongyan Li, Avi Natan, Yuanyue Liu, Daxian Cao, and Hongli  
Zhu\**

## Supporting Information

### Mass Transfer and Reaction Kinetic Enhanced Electrode for High-Performance Aqueous Flow Batteries

Alolika Mukhopadhyay,<sup>1£</sup> Yang Yang,<sup>1£</sup> Yifan Li,<sup>2</sup> Yong Chen,<sup>1</sup> Hongyan Li,<sup>1</sup> Avi Natan,<sup>1</sup> Yuanyue Liu,<sup>2</sup> Daxian Cao,<sup>1</sup> Hongli Zhu<sup>1\*</sup>

<sup>1</sup>Department of Mechanical and Industrial Engineering, Northeastern University, 360 Huntington Avenue, Boston, Massachusetts 02115, United States

<sup>2</sup>Department of Mechanical Engineering, The University of Texas at Austin, 110 Inner Campus Drive, Austin, TX 78705, United States

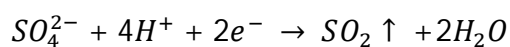
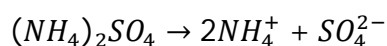
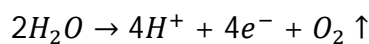
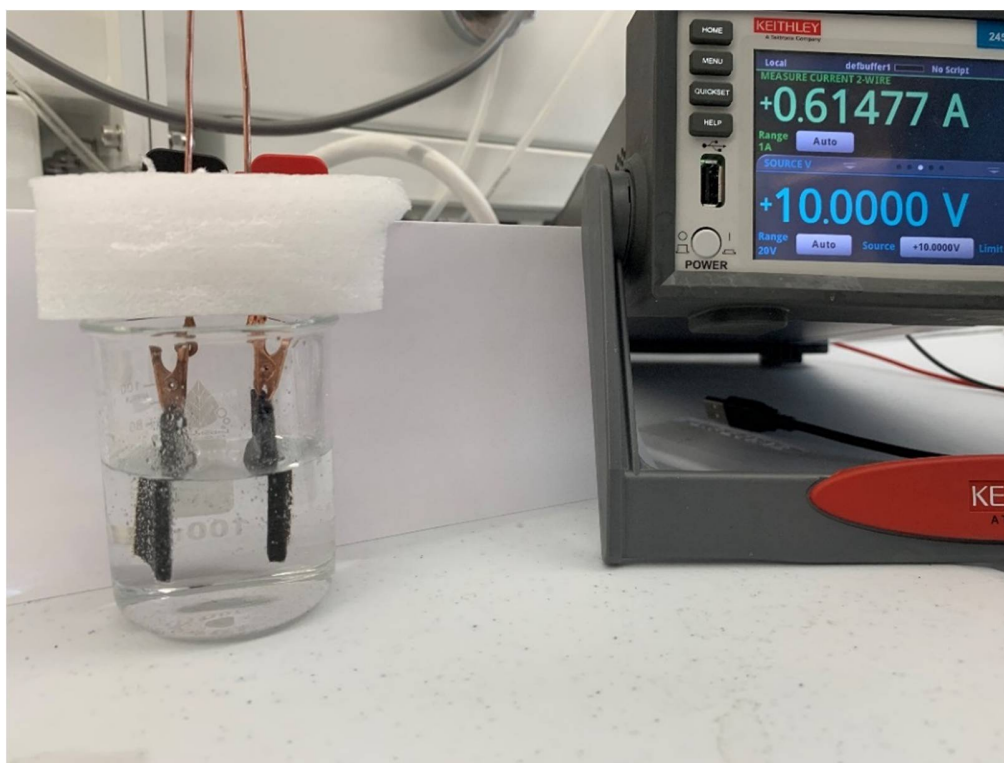
\*: Corresponding author: Dr. Hongli Zhu. E-mail: [h.zhu@neu.edu](mailto:h.zhu@neu.edu)

£: These authors contributed equally to this work.

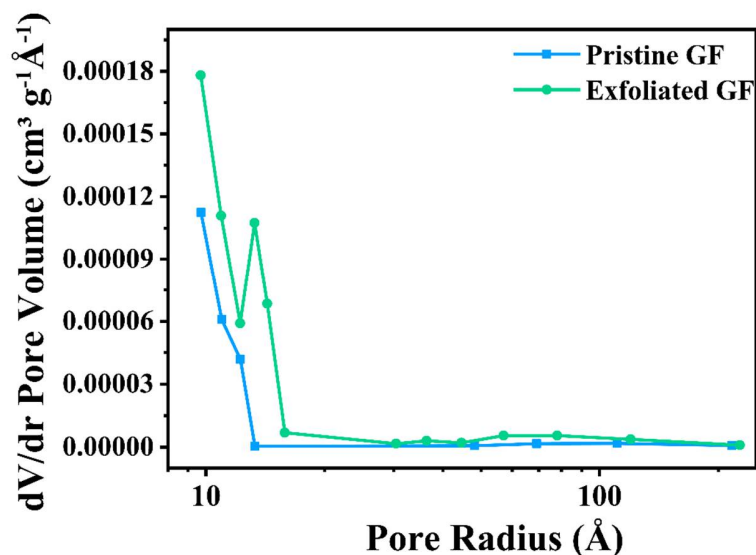
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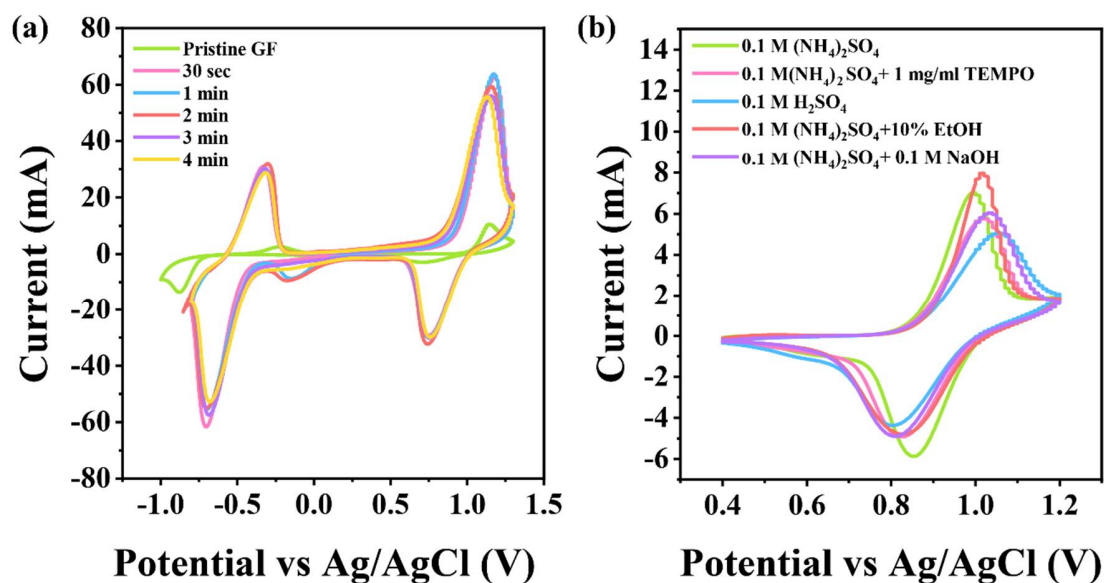
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The charge density difference between neutral and +1/-1 charged OH and H terminated graphene.
- Supplementary Figure 14** (a) Raman spectra and (b) I<sub>D</sub>/I<sub>G</sub> ratio of E-GF samples exfoliated for 30 sec, 1 min, 2 min, 3 min, and 4 min.
- Supplementary Figure 15** (a) Digital photographs of exfoliated graphite felt for different times ranging from 30 sec to 4 min highlighting the wettability compared to the pristine graphite felt in deionized water. (b) The sheet resistance of exfoliated graphite felts for different times ranging from 30 sec to 4 min.
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**Figure S1** Picture of exfoliation set up during exfoliation of the graphite felt electrode



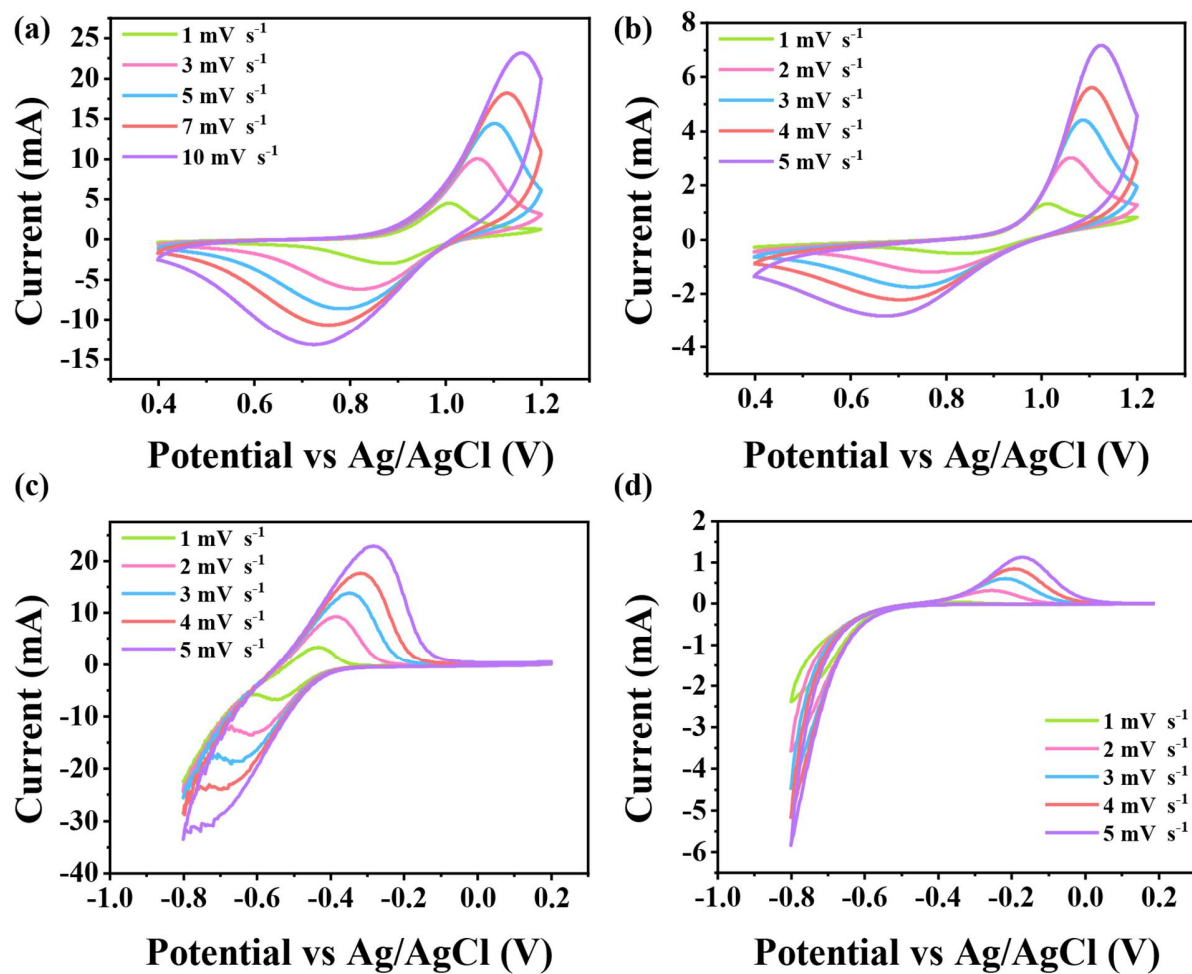
**Figure S2** The pore size distribution of the pristine graphite felt and exfoliated graphite felt using Brunauer-Emmett-Teller.



**Figure S3** (a) CV curves of E-GF electrodes exfoliated for various times ranging from 0 sec to 4 min for  $VO_2^+ / VO^{2+}$  and  $V^{3+} / V^{2+}$  redox couples. (b) CV curves of 1 min E-GF electrodes exfoliated in various electrolyte solutions.

*Table S 1: Summary of critical cyclic voltammetry parameters for exfoliated graphite felts for different times ranging from 1 min to 4 min*

Critical parameters	Different exfoliation times				
	30 sec	1 min	2 min	3 min	4 min
<b>For VO<sub>2</sub><sup>+</sup>/VO<sup>2+</sup> redox couple</b>					
Peak separation (V)	0.4289	0.42529	0.419	0.4045	0.3745
Oxidation onset potential (V)	0.94	0.926	0.884	0.898	0.8758
Reduction onset potential (V)	0.9733	0.992	0.986	0.991	0.9927
Oxidation peak Current (mA)	62.7	63.8	59.1	55.8	54.4
Reduction peak Current (mA)	29.15	29.97	30.04	30.14	29.94
<b>For V<sup>3+</sup>/V<sup>2+</sup> redox couple</b>					
Peak separation (V)	0.3775	0.38586	0.3997	0.366	0.36874
Oxidation onset potential (V)	-0.573	-0.5377	-0.5389	-0.54	-0.5089
Reduction onset potential (V)	-0.4545	-0.429	-0.422	-0.4294	-0.4163
Oxidation peak Current	31.19	29.70	30.73	30.18	28.83
Reduction peak Current	61.75	55.14	54.78	53.32	52.68



**Figure S4** CV curves of (a) T-GF electrode and (b) A-GF electrode at different scan rates ranging from 1 to 10  $mV s^{-1}$  for  $VO_2^+/VO_2^{2+}$  redox couple. CV curves of (c) T-GF electrode and (d) A-GF electrode at different scan rates ranging from 1 to 10  $mV s^{-1}$  for  $V^{3+}/V^{2+}$  redox couple.

**Randles–Sevcik** equation describes the effect of scan rate on the peak current  $I_p$  and can be represented as follows

$$I_p = 0.4463 nFAC \left(\frac{nf\vartheta d}{RT}\right)^{\frac{1}{2}} \dots\dots\dots \text{Equation S 1}$$

Where,

$I_p$  = Peak current in amps

$n$  = Number of electrons transferred in the redox event (1 for  $\text{VO}_2^+/\text{VO}^{2+}$  and  $\text{V}^{3+}/\text{V}^{2+}$  redox couples)

$A$  = Electrode area in  $\text{cm}^2$

$F$  = Faraday Constant in  $\text{C mol}^{-1}$

$D$  = Diffusion coefficient in  $\text{cm}^2 \text{s}^{-1}$

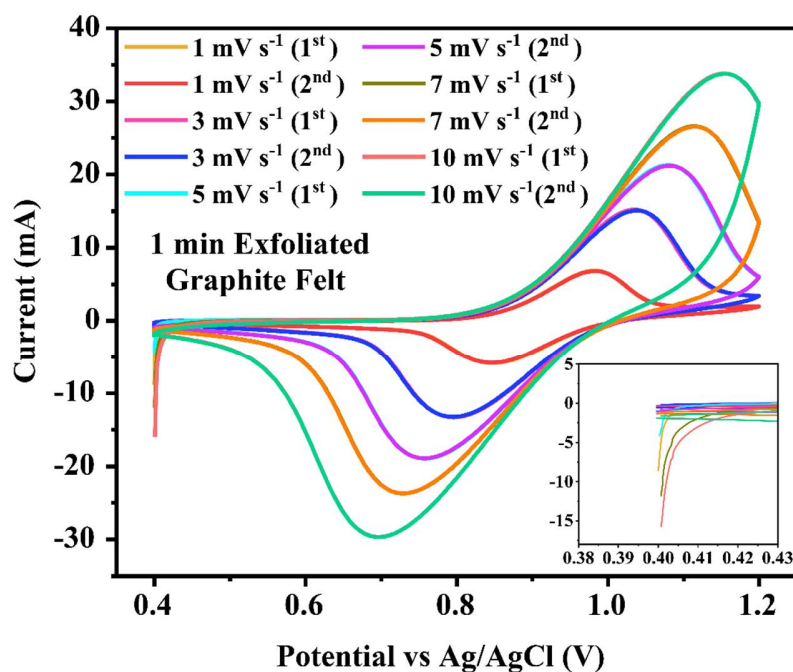
$C$  = Concentration in  $\text{mol cm}^{-3}$

$v$  = Scan rate in  $\text{V/s}$

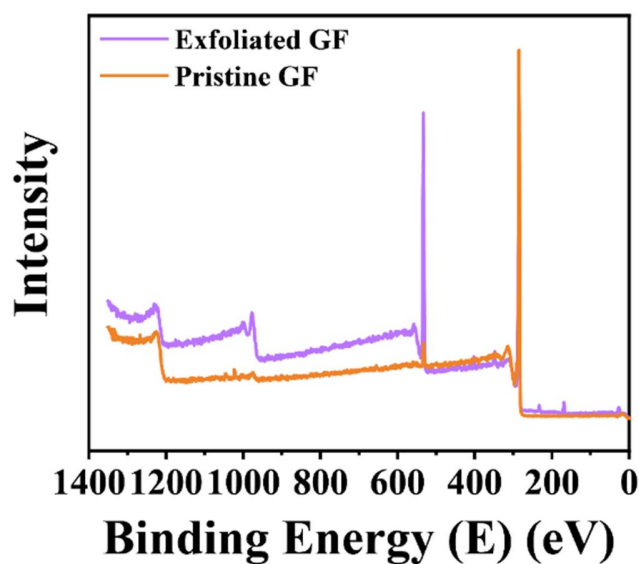
$R$  = Gas constant in  $\text{J K}^{-1} \text{mol}^{-1}$

$T$  = Temperature in  $\text{K}$

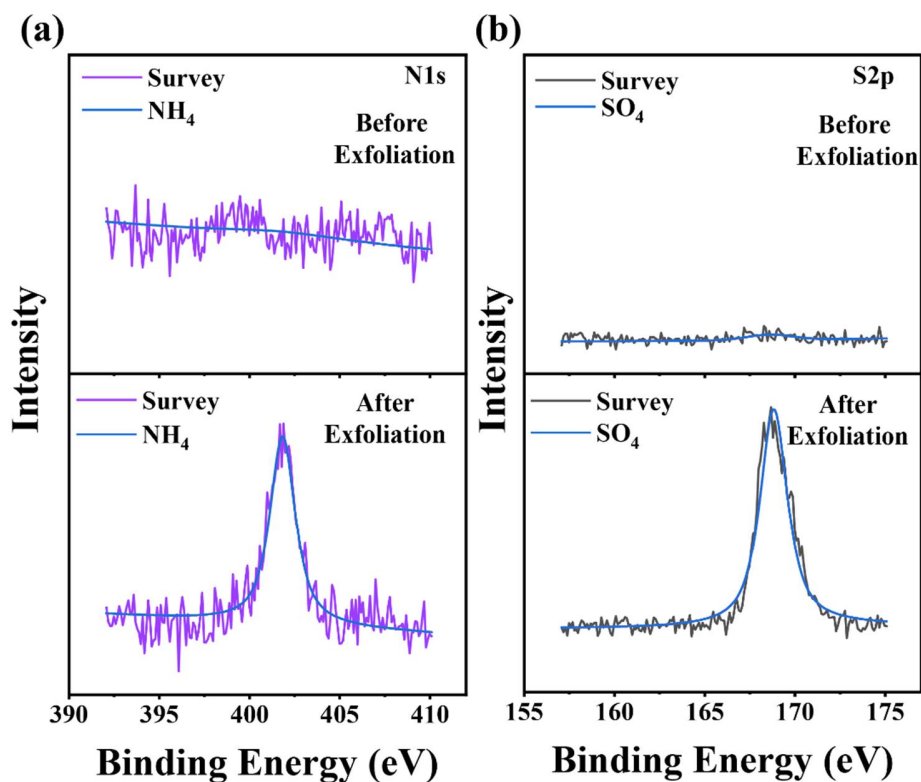




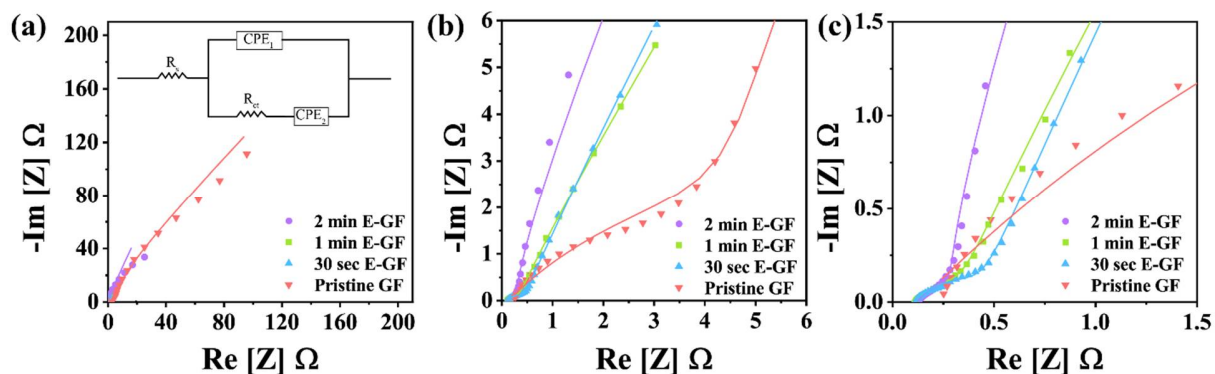
**Figure S5** 1<sup>st</sup> and 2<sup>nd</sup> cycle of the cyclic voltammetry curves of exfoliated 1 min exfoliated graphite felt electrode in 0.1 M VOSO<sub>4</sub> in 3 M H<sub>2</sub>SO<sub>4</sub> for the VO<sub>2</sub><sup>+</sup>/VO<sub>2</sub><sup>2+</sup> redox couple at different scan rates (1 mV s<sup>-1</sup> to 10 mV s<sup>-1</sup>.)



**Figure S6** Wide range XPS spectra of pristine and exfoliated graphite felt.

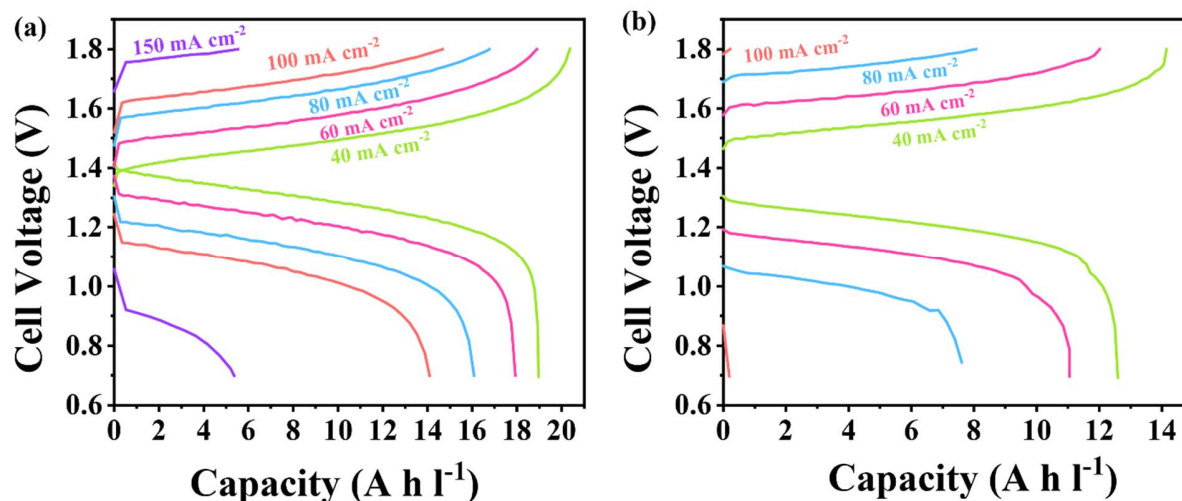


**Figure S7** High-resolution XPS spectra of pristine and exfoliated graphite felt for (a) N1s (b) S2p.

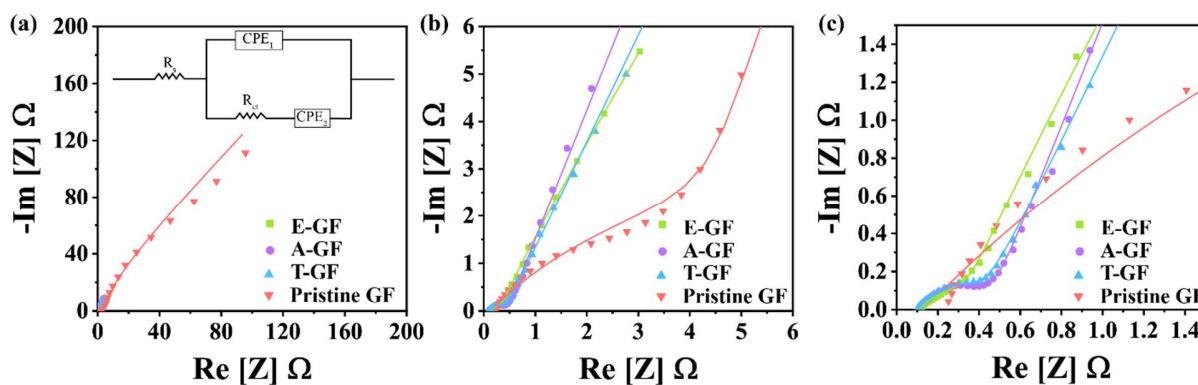


**Figure S8** Nyquist plots of VRFB employing E-GF electrodes with varying exfoliation (0 sec to 2 min). (a) Original plot. The inset shows the equivalent circuit for EIS fitting. (b) Zoomed in part

of (a). (c) Zoomed in part of (b) to show the semicircles. The symbols represent the measured data, and the line represents the corresponding fitting.

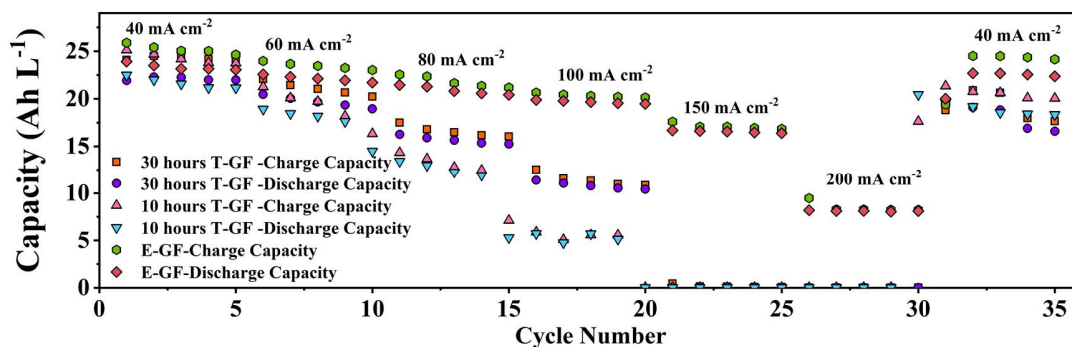


**Figure S9** Charge-discharge profiles of E-GF electrodes exfoliated for (a) 30 sec and (b) 2 min at current densities of 40, 60, 80, 100, and 150 mA cm<sup>-2</sup>.

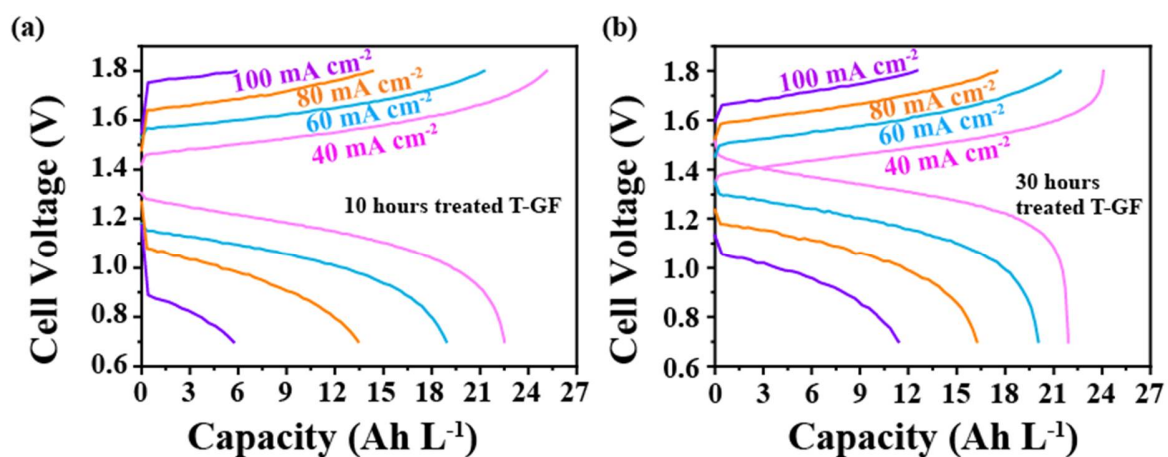


**Figure S10** Nyquist plots of VRFB employing E-GF electrodes exfoliated for various-electrodes.

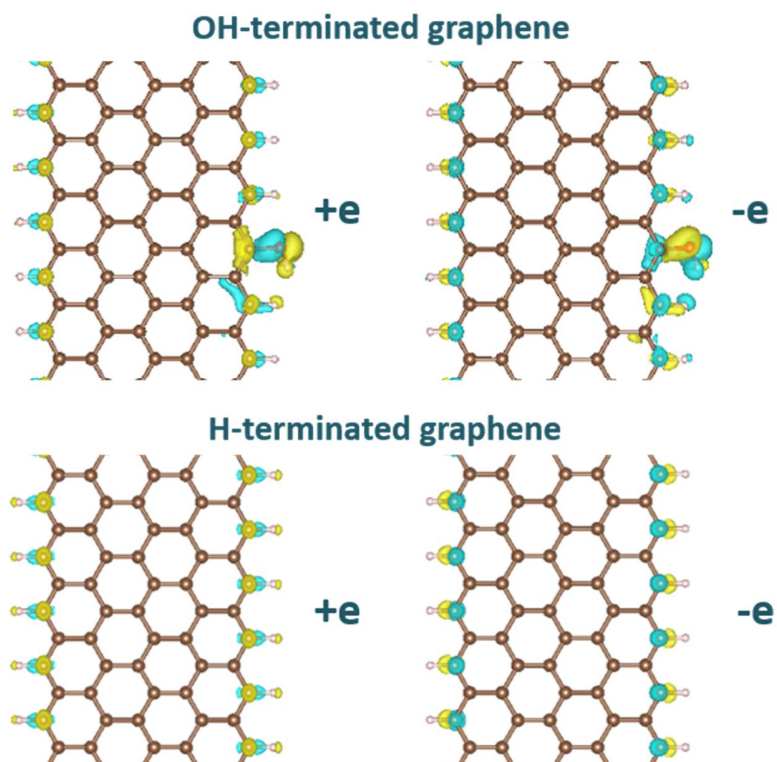
(a) Original plot. The inset shows the equivalent circuit for EIS fitting. (b) Zoomed in part of (a). (c) Zoomed in part of (b) to show the semicircles. The symbols represent the measured data, and the line represents the corresponding fitting.



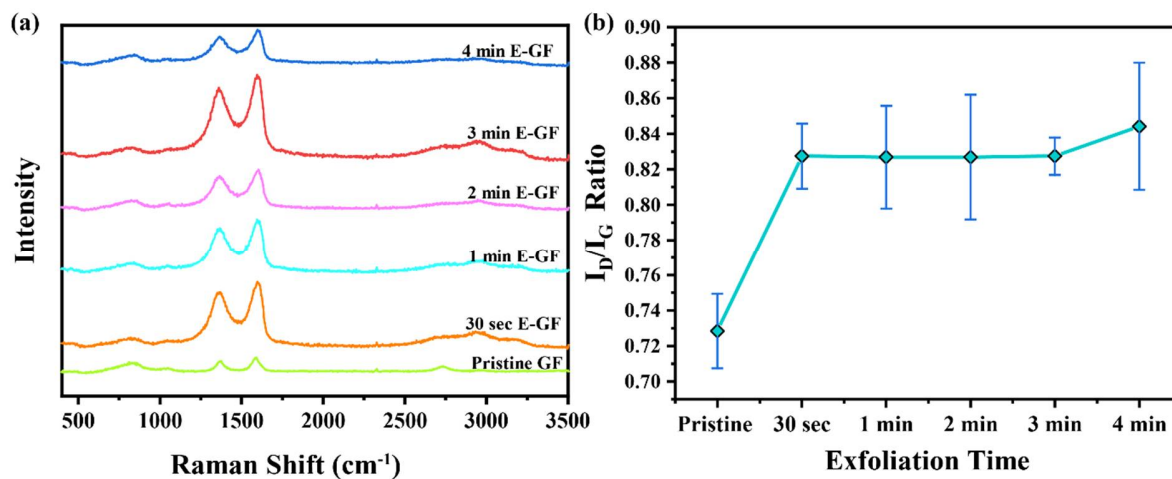
**Figure S11** Rate Performance of E-GF, 30 hours treated T-GF, and 10 hours treated T-GF electrodes at different current densities of 40, 60, 80, 100, 150, and 200 mA cm<sup>-2</sup>.



**Figure S12** Charge-discharge profiles of T-GF electrodes treated for (a) 10 hours and (b) 30 hours at 400°C in the air at current densities of 40, 60, 80, and 100 mA cm<sup>-2</sup>.



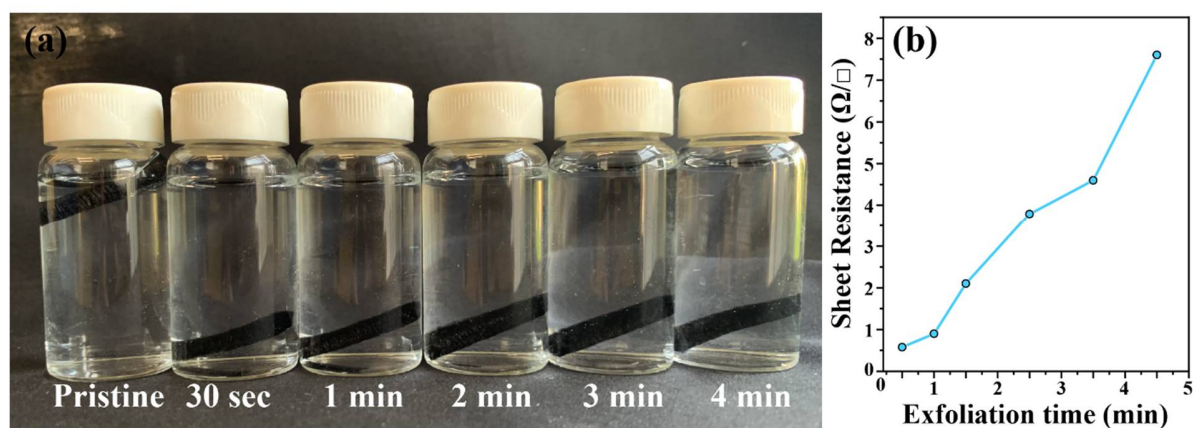
**Figure S13** The charge density difference between neutral and +1/-1 charged OH and H terminated graphene.



**Figure S14** (a) Raman spectra and (b)  $I_D/I_G$  ratio of E-GF samples exfoliated for 30 sec, 1 min, 2 min, 3 min, and 4 min.

**Table S 2** Sheet resistance measurement

Samples	Sheet Resistance ( $\Omega/\square$ )
Pristine graphite felt	0.575
30 sec exfoliated graphite felt	0.9
1 min exfoliated graphite felt	2.1
2 min exfoliated graphite felt	3.79
Acid-treated graphite felt	0.66
Thermally treated graphite felt	2.5



**Figure S15** (a) Digital photographs of exfoliated graphite felt for different times ranging from 30 sec to 4 min highlighting the wettability compared to the pristine graphite felt in deionized water. (b) The sheet resistance of exfoliated graphite felts for different times ranging from 30 sec to 4 min.