Electronic Supporting Information for

Graphene Quantum Dots Decorated Carbon Electrodes for Energy Storage in Vanadium Redox Flow Batteries

Table S1. The ASR magnitudes and fitted parameters for the spectra shown in Figure 3.

Component \ Electrode	**QD-1**	**QD-2**	**QD-3**	**Untreated**
ASR charge transfer (Ω cm^2)	4.09	3.46	3.67	4.56
ASR diffusion (Ω cm^2)	1.20	0.60	0.74	1.76
ASR ohmic (Ω cm^2)	0.95	0.95	0.93	0.93
Double layer capacitance (mF)	2.62	3.39	2.98	1.98
Exchange current density (μA cm^{-2})	0.25	0.22	0.23	0.37
Diffusion layer thickness (μm)	7.90	8.17	8.18	6.36
P (CPE exponent)	0.90	0.89	0.89	0.93
***f* (Scale factor)**	0.03	0.05	0.05	0.03

Table S2. The symbols and their corresponding magnitudes used in the mathematical model.

Symbol	Item	Value	Units
a	Diffusion layer thickness	*fit*	cm
A	Geometric surface area	5	cm^2
A_t	Total surface area of electrode	*fit*	cm^2
b	Electrode thickness (compressed)	0.2	cm
C_{dl}	Specific double layer capacitance	2×10^{-5}	F cm^{-2}
C_o	Concentration of oxidized species	0.0005	mol cm^{-3}
C_R	Concentration of reduced species	0.0005	mol cm^{-3}
D_O	Diffusivity of oxidized species	0.57×10^{-6} [60]	cm^2 s^{-1}
D_R	Diffusivity of reduced species	1.1×10^{-6} [60]	cm^2 s^{-1}
f	Scale factor	*fit*	---
F	Faraday's constant	96,485	C mol^{-1}
i_0	Exchange current density	*fit*	A cm^{-2}
j	Imaginary unit	$\sqrt{-1}$	---
ω	Angular frequency	---	rad s^{-1}
P	Constant phase element exponent	*fit*	---
Q	CPE capacitance	$A_t \times C_{dl}$	F
ρ_1	Specific ionic resistance within the electrode	3.24 [61]	Ω cm
ρ_2	Specific electronic resistance of the electrode	0.3 [manufacturer spec]	Ω cm
R	Gas constant	8.314	J K^{-1} mol^{-1}
T	Temperature	303.15	K
Z_p	Impedance of a single electrode	---	Ω
$Z_i^{'}$	Real component of Z at frequency i	---	Ω
$Z_i^{''}$	Imaginary component of Z at frequency i	---	Ω

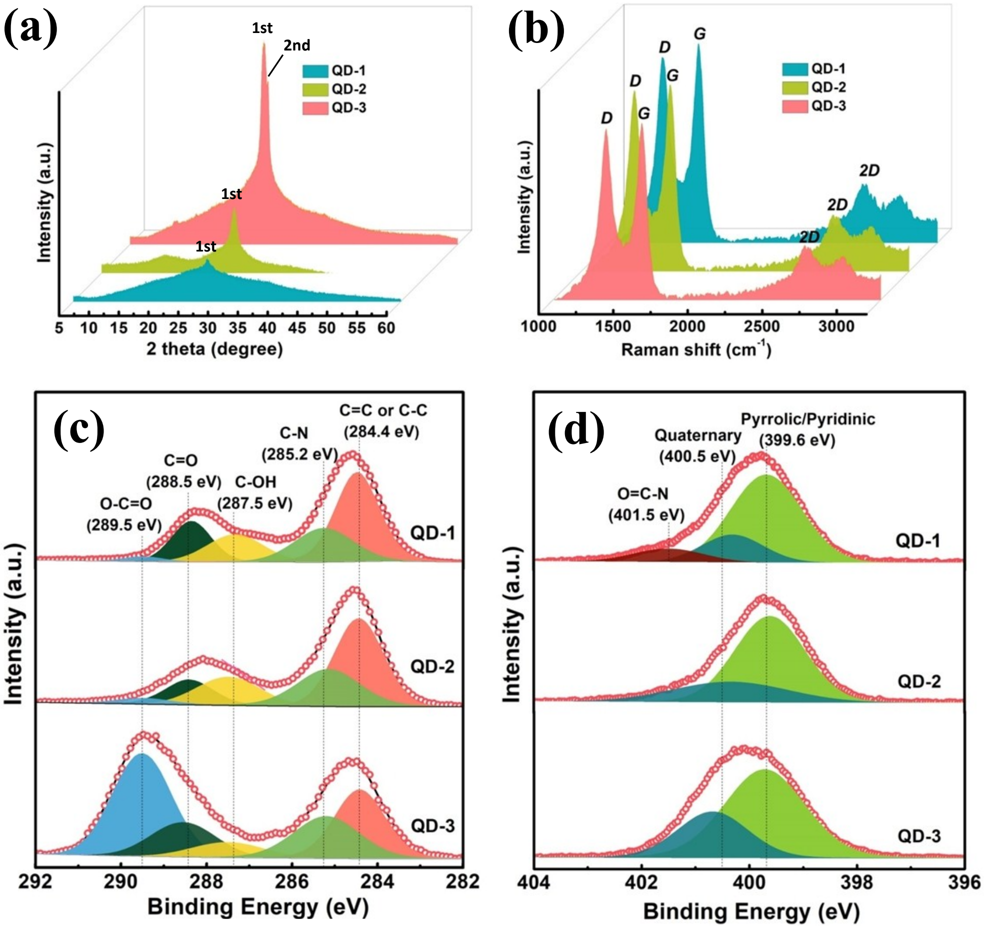

Figure S1. (a) XRD patterns and (b) Raman spectra of QD-1, QD-2, and QD-3 samples. XPS (c) C 1s and (d) N 1s peaks of QD-1, QD-2, and QD-3 samples deconvoluted by a multiple Gaussian function [27].

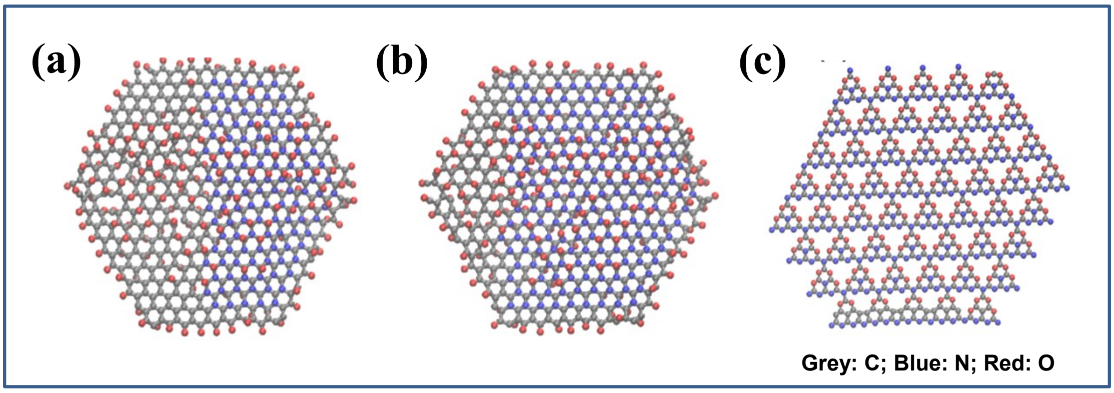

Figure S2. The atomic configurations of (a) QD-1, (b) QD-2, and (c) QD-3 samples, simulated by the Molecular Dynamics software [27]. The simulations were carried out using the software package Large-scale Atomic/Molecular Massively Parallel Simulator (LAMMPS) with the Reax force field.

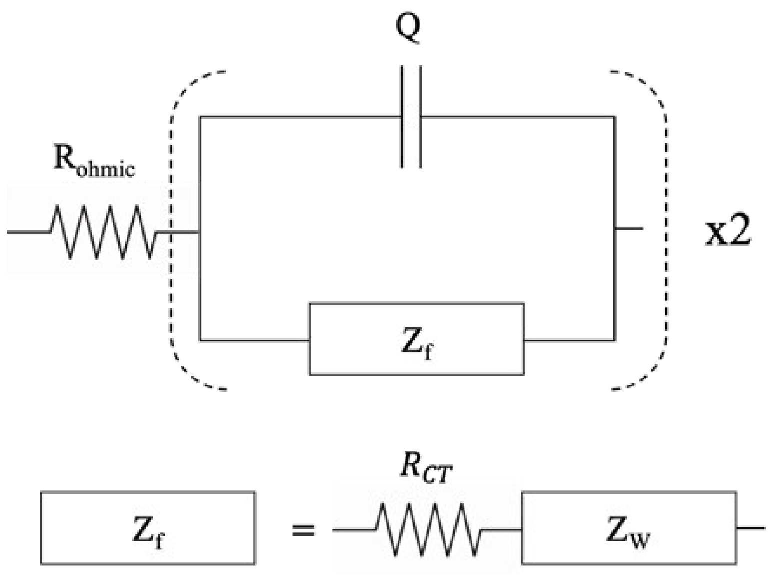

Figure S3. Equivalent circuit proposed for describing the electrochemical impedance behavior of the symmetric cell VRFBs, where R_{ohmic}, R_{CT}, Z_{f}, Z_{warburg} and Q components represent ohmic resistance, charge transfer resistance, faradaic impedance, Warburg impedance, and constant phase element capacitance, respectively.

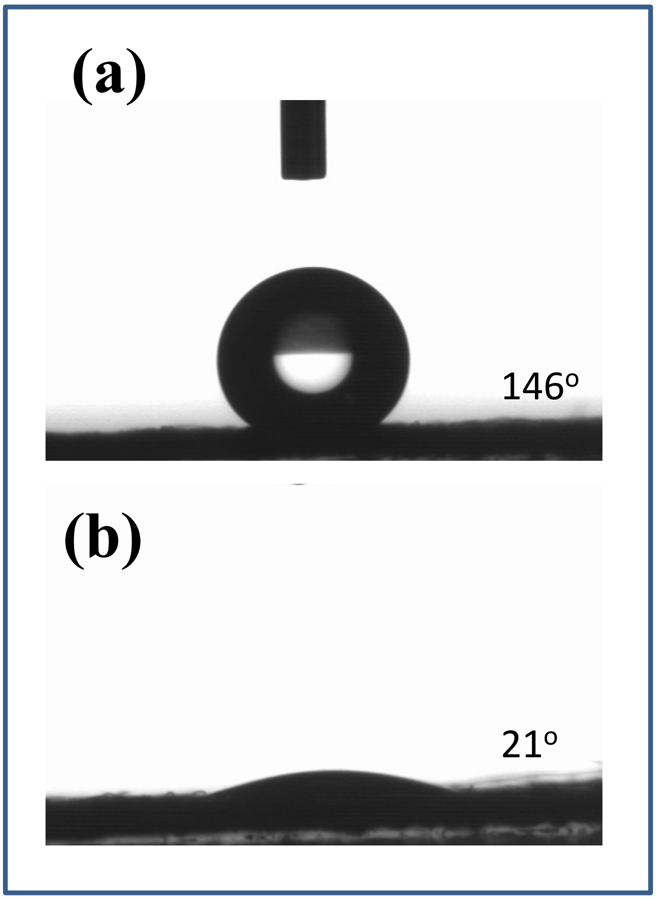

Figure S4. Cross-sectional views of water droplets on (a) pristine GF and (b) QD-2 samples.

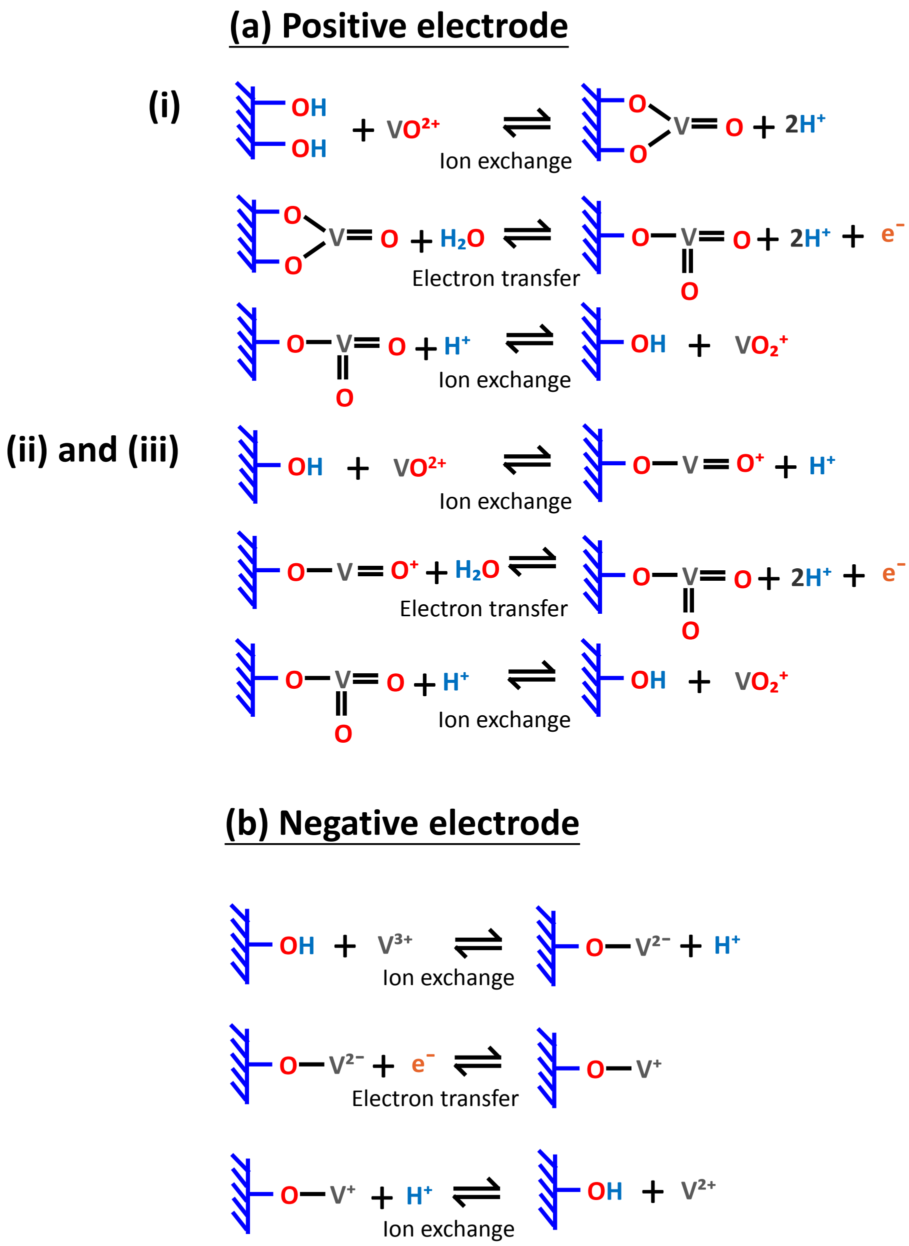

Figure S5. Two sets of possible surface reaction steps at (a) positive and (b) negative GQD/GF electrodes of VRFBs [14,44].

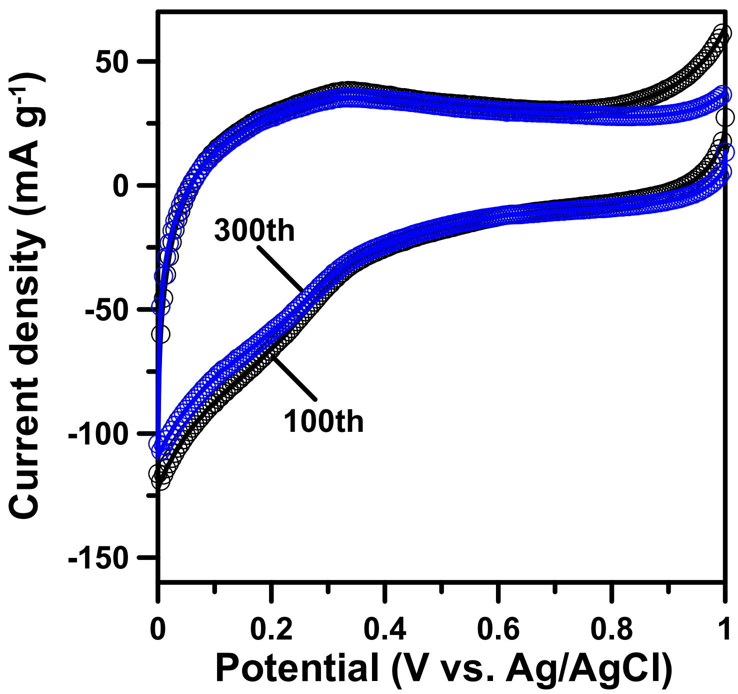

Figure S6. Typical CV profiles of QD-2 electrode in 1 M sulfuric acid under nitrogen steady flow at 50 mV s^{-1}.

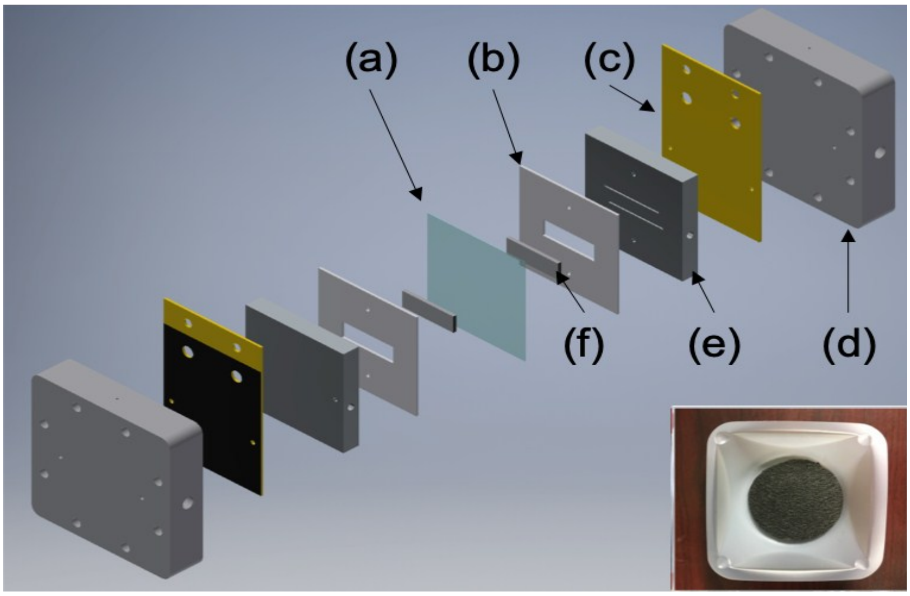

Figure S7. Schematic of the VRFB equipped with GQD/GF electrodes (including positive and negative electrodes). The components are (in clockwise order): (a) membrane, (b) gasket, (c) current collector, (d) end plate, (e) graphite plate, and (f) electrode. The inset shows the GQD/GF electrode sheet.

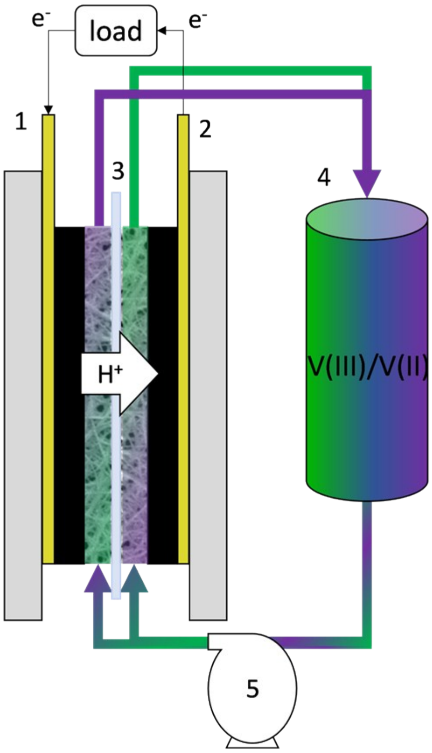

Figure S8. Schematic of symmetric cell configuration, where (1) and (2) are respectively the cathode and anode in this schematic (though these can be switched for the same operation), (3) is the point where the reference electrode contacts the cell, (4) is the electrolyte reservoir (50% SoC), and (5) is the pump.

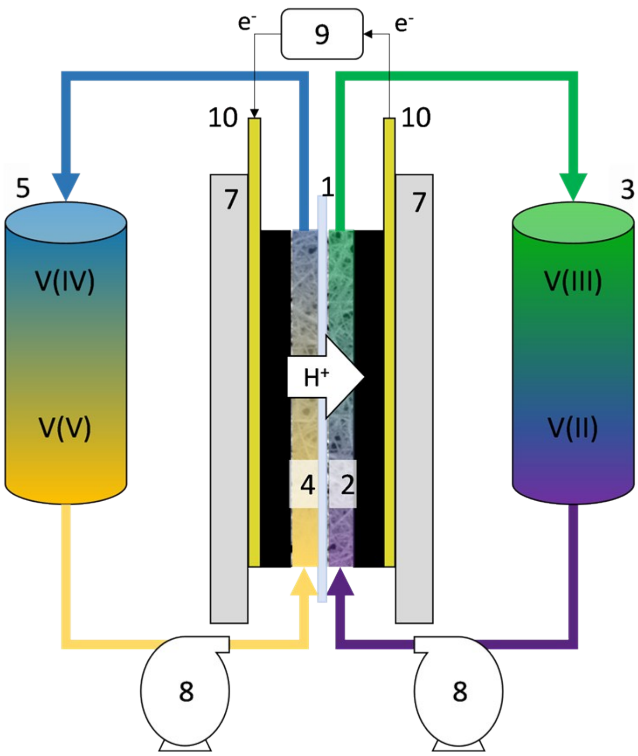

Figure S9. Schematic of full cell configuration in discharge mode, where the components include (1) membrane/separator, (2) anode, (3) anolyte tank, (4) cathode, (5) catholyte tank, (6) graphite flow plates, (7) aluminum end plates, (8) pumps, (9) load (during discharge), and (10) current collectors.