Supporting Information

Hydrophilic Anthraquinone-Substituted Polymer: Its Environmentally Friendly Preparation and Efficient Charge/Proton-Storage Capability for Polymer–Air Secondary Batteries

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Supplementary Method

1. Electrode preparation

The polymer/carbon composite electrodes were prepared by coating the slurry of the polymer single-walled carbon nanotube (SWNT), polyvinylidene difluoride (5:5:1 in w/w/w), and *N*-methyl pyrrolidone (NMP) onto glassy carbon substrates. The polymer carbon composite was coated on a glassy carbon plate with a thickness of ca. 10 μm. The mass loading of the polymer was adjusted to ca. 1.0 mg.

2. Electrochemical measurement

A tailor-made glass cell (20 cm^2 electrolyte, Watanabe Kagaku Co.) was employed as the electrochemical cell.

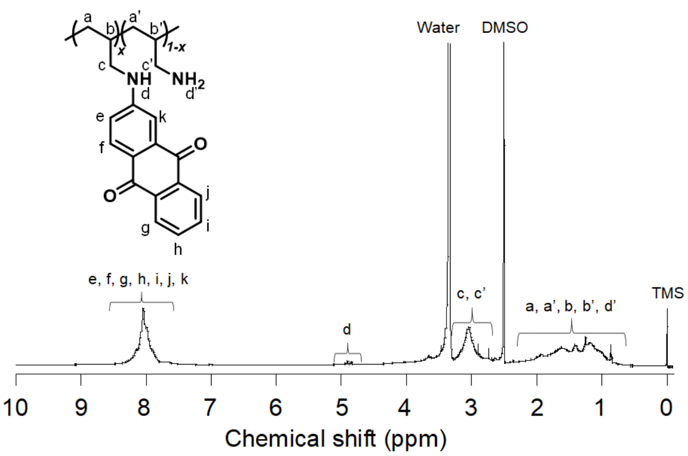

Figure S1. 500 MHz ^{1}H NMR spectrum (dimethyl sulfoxide-d_6) of anthraquinone substituted poly(allylamine).

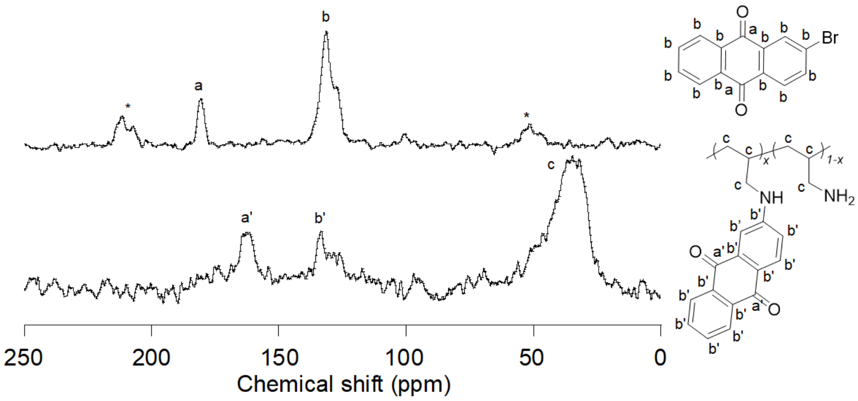

Figure S2 Solid state ^{13}C-NMR spectra of anthraquinone substituted poly(allylamine) (* spinning side band).

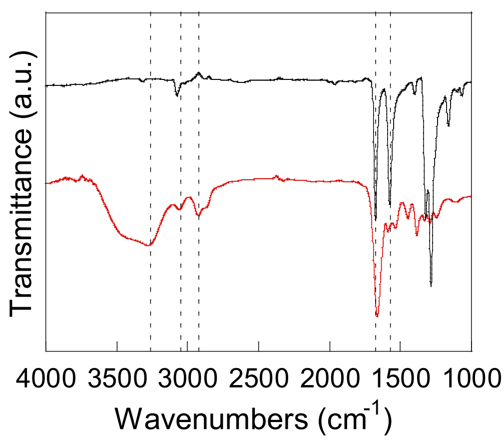

Figure S3 IR spectra of bromoanthraquinone (black) and anthraquinone-substituted poly(allylamine) (red).

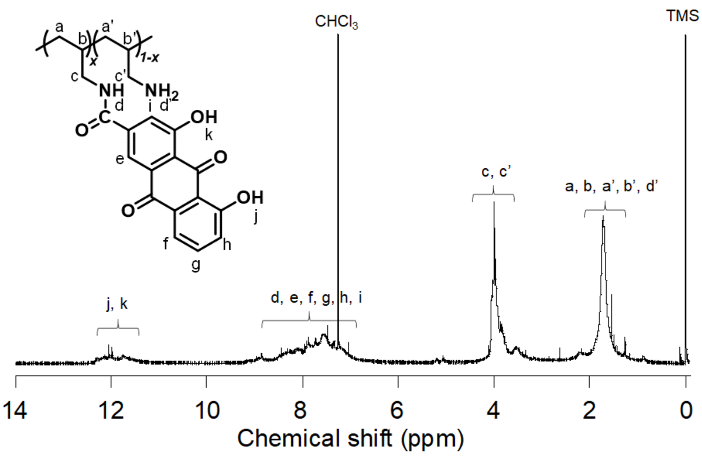

Figure S4. 500 MHz ^{1}H NMR spectrum ($CDCl_3$) of PDHA.

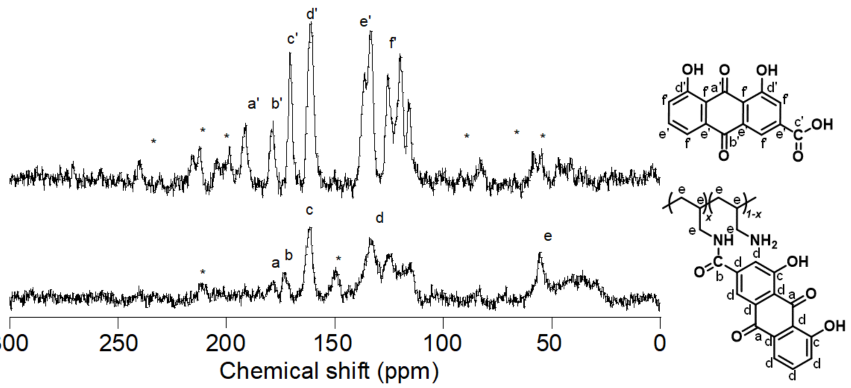

Figure S5. Solid state ^{13}C-NMR spectra of a) bromoanthraquinone and b) PDHA (* Spinning side band).

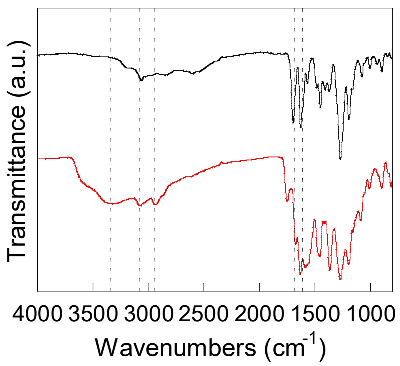

Figure S6. IR spectra of 4,5-dihydroxyanthraquinone-2-carboxylic acid (black) and PDHA (red).

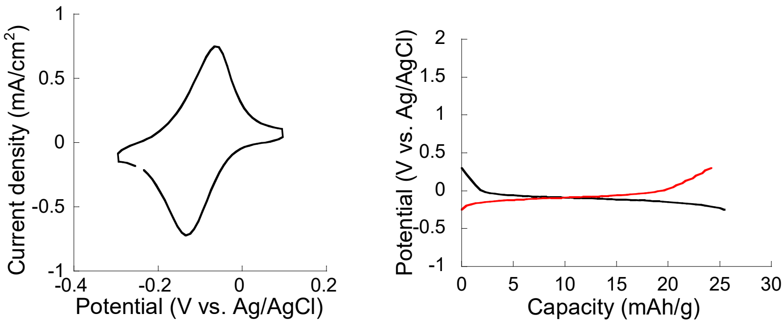

Figure S7. a) Cyclic voltammogram and b) Charging (black) and discharging (red) curves of the anthraquinone substituted poly(allylamine)/SWNT composite electrode in 0.5 M H_2SO_4 aqueous solution at 10 C.

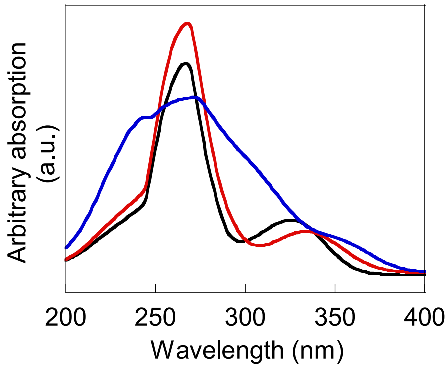

Figure S8. The UV-vis absorption spectra (arbitrary absorption) of anthraquinone (0.1 mM) (black), poly(vinylanthraquinone) (0.1 mM) (red), and anthraquinone substituted poly(allylamine) (0.1 mM) (blue) in NMP.

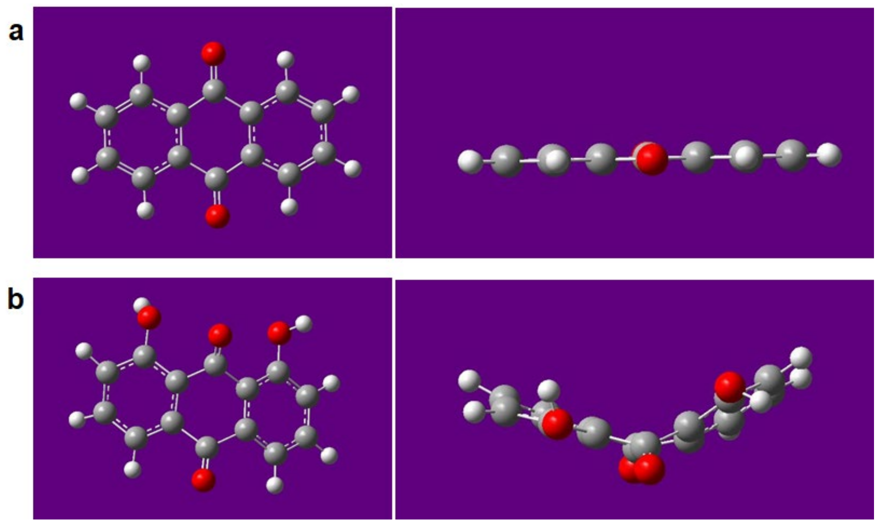

Figure S9. UM06/6-31G(d,p) optimized structures of (a) anthraquionone and (b) DHA.

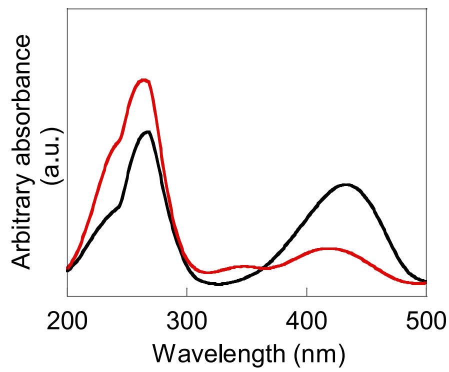

Figure S10. The UV-vis absorption spectra (arbitrary absorption) of DHA (0.1 mM) (black) and PDHA (0.1 mM) (red) in NMP.

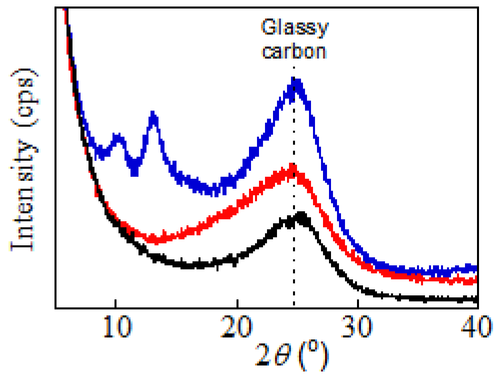

Figure S11. XRD patterns of the thin films of poly(vinylanthraquinone) (blue) and PDHA (red), and glassy carbon substrate (black). These thin films are formed on the glassy carbon substrate.

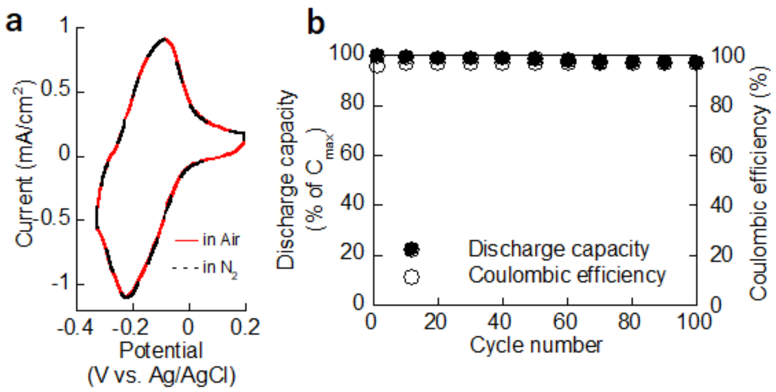

Figure S12. a) Cyclic voltammograms of the PDHA/SWNT composite electrode in 0.5 M H_2SO_4 aqueous solution in air (red solid line) and in nitrogen (black dotted line) at a scan rate of 10 mV/s. b) Capacity retention for 100 cycles on galvanostatic charge and discharge at 60 C.

Table S1 Characteristics of other organic-air battery concepts

Reference No.	Anode-active Polymer (conductive additives)	Cathode-active Catalyst (conductive additives)	Electrolyte	Voltage (V)	Capacity loss (100 cycles)
1	Poly(vinylanthraquinone) (ca. 100 nm layer)	VGCF, MnO_2, and PVdF	30 wt% KOH aqueous solution (pH 14)	0.5	-13%
2	poly(dianthraquinone-substituted norbornene) (ca. 50 nm layer)	VGCF, MnO_2, and PVdF	10 M NaOH aqueous solution	0.7	-3%
3	poly(1,4-anthraquinone) (P14AQ) in situ polymerized on carbon nanotubes (CNTs)	spinel cobalt manganese oxide supported on CNTs	6.0 M KOH aqueous solution	0.8	-5%
4	poly(2,5-dihydroxy-1,4-benzoquinone-3,6-methylene), single-wall carbon nanotube, and poly(vinylidene fluoride)	Pt/C	H_2SO_4 aqueous solution (pH 1)	0.4	-3%
5	a conducting redox polymer based on a trimer of EPE (E=3,4-ethylenedioxythiophene; P=3,4-propylenedioxythiophene) and a naphthoquinone (NQ) pendant group	Pt/C	H_2SO_4 aqueous solution (pH 1)	0.5	-2%
6	naphthoquinone substituted poly(allylamine), single-wall carbon nanotube, and poly(vinylidene fluoride)	Pt/C	H_2SO_4 aqueous solution (0.05 M H_2SO_4)	0.8	-7%
This	1,8-dihydroxyanthraquinone-	Pt/C	H_2SO_4	1.1	-2%

work	substituted poly(allylamine), single-wall carbon nanotube, and poly(vinylidene fluoride)		aqueous solution (0.5 M H_2SO_4) in air		

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