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Supporting Information

High Power Zinc Iodine Redox Flow Battery with Iron Functionalized Carbon Electrodes Abena A. Williams,^a Robert K. Emmett^b and Mark E. Roberts^{a,*}

Materials & Methods

Electrode Fabrication. Two types of multiwalled carbon nanotubes (NanoTech Labs) namely Fe-CNT (Mgrade, diameter = 70-80 nm, length = 10 μ m) and C-CNT (C-grade, diameter = 50 nm, length = 10 μ m) were sonicated in an aqueous solution of 0.052 M sodium dodecyl sulfate (Sigma Aldrich) for approximately 30 min to disperse the carbon nanotubes (0.60 mg/mL). The suspension was vacuum filtered with a nylon membrane (0.47 um). The filtrate was washed with deionized water several times to remove the residual sodium dodecyl sulfate and the resulting film shaved off the nylon membrane to produce carbon nanotube paper electrodes. The electrodes were oven dried overnight to remove excess water and then soaked in the redox electrolytes before testing.

Separator preparation. Nafion-212 membranes (0.05mm, Alfa Aesar) was activated by refluxing at 90 °C for 1 h each with DI water, 3 v% hydrogen peroxide (30% Fisher), DI water, 0.5 M sulfuric acid (98% Fisher), DI water, and 1 M potassium chloride (Alfa Aesar). Activated membranes were stored in 1M potassium chloride until use.

Electrolyte Preparation. A 1 M electrolyte was used for all electrochemical characterization. The electrolyte consists of 1 M potassium iodide (Acros Organics) and 0.5 M zinc bromide (Alfa Aesar) in a 1 M potassium chloride (Fisher Scientific) solution. 250 mL of the electrolyte was prepared from a 1 M KCl solution in deionized water by adding 41.5 g KI and 28.15 g ZnBr₂. The electrolyte was stirred at 50°C for about 15 min or until everything dissolved.

Testing Configuration and Electrochemical Characterization. Cyclic voltammetry (CV) tests were performed in a three-electrode cell using a VersaSTAT 4 Potentiostat (Princeton Applied Research) to investigate the electrochemical performance of the electrolyte system on both electrode types. The reference electrode was Ag/AgCl in 3M NaCl, the counter electrode was a graphite electrode, and the working electrode was 15 mg of 2 mm x 1 mm CNT pasted on a glassy carbon electrode (GCE) with carbon conductive adhesive. Prior to testing, the working electrode was soaked 1 M electrolyte for about 2 h. Tests were conducted from -1.5 V to 1.5 V versus Ag/AgCl at varying scan rates.



Figure S1. Nyquist plot from electrochemical impedance spectroscopy measurements on batteries with Fe-CNT (solid lines) and C-CNT (dashed lines) electrodes.



Figure S2: 3-electrode Cell.

A zinc iodine redox flow battery was assembled using the micro flow cell from Electrocell. The twocompartment cell was assembled with stainless steel end plates, PTFE end frames, rubber gaskets, graphite current collectors, PTFE flow frames, PVDF turbulence mesh and a 3 x 3 cm electrode surface area with the activated nafion membrane separating both half cells. The previously soaked CNT paper electrodes were used as both cathode and anode and the same electrolyte (1 M) was used as both the catholyte and anolyte. The electrolytes were circulated through the cell using a dual channel peristaltic pump (EQ-BK-380-2) from MTI Corps via viton Tubing (F-5500-A) from US Plastics.

Prior to electrochemical analysis of the redox flow battery, the electrolyte was pumped through the cell for 10 min to ensure there was no leaking and electrodes were saturated with electrolyte. The electrodes were conditioned under zero flow by CV cycling at 10 mV/s and 5 mV/s from -1.6 V to 1.6 V and then continuous 1 h charge and then discharge at 100 mA at 30 mL/min until stable behavior was observed. The electrochemical performance of the electrodes was analyzed by galvanostatic charge-discharge (GCD) tests at various current densities, polarization analysis at varying flowrates and electrochemical impedance spectroscopy (EIS). GCD and EIS tests were controlled by the Arbin MSTAT21044 Potentiostat and Gamry Instruments Reference 600 Potentiostat, respectively.





Figure S4: Pilot scale zinc iodine redox flow battery.