

## Supporting Information

### **An ionic aqueous pseudocapacitor system: electroactive ions in both salt-electrode and redox-electrolyte**

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### **Experimental section**

Electrode preparation and electrochemical measurement. Slurries containing chloride salts ( $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ ,  $\text{CoCl}_2 \cdot 6\text{H}_2\text{O}$ ,  $\text{NiCl}_2 \cdot 6\text{H}_2\text{O}$ , or  $\text{CuCl}_2 \cdot 2\text{H}_2\text{O}$ ), carbon black, and poly(vinylidene fluoride) (PVDF) in a weight ratio of 70:20:10 were prepared by mixing these materials and then dispersing them in N-methyl-2-pyrrolidone (NMP). Electrodes were prepared by spreading the slurry on a nickel foam current collector with an area of  $1 \times 1 \text{ cm}^2$ . The electrode was dried at  $80 \text{ }^\circ\text{C}$  for 24h and finally pressed at 10 MPa. The total weight of active component in an electrode is fixed to be approximately 3 mg.

Electrolytes are 2M KOH or mixed redox-electrolyte including 2 M KOH and different concentrations of  $\text{K}_3\text{Fe}(\text{CN})_6$  (0.05, 0.1, 0.2, and 0.3 M). All electrochemical experiments were carried out using a classical three-electrode cell configuration with the saturated calomel electrode (SCE) as the reference electrode, and Pt wire as a counter electrode. The cyclic voltammograms (CV), and galvanostatic charge-discharge measurements were carried out by an electrochemical workstation (CHI 660D). The applied potential ranges were  $-0.1$ - $0.45 \text{ V}$  for  $\text{CoCl}_2$  electrode,  $0$ - $0.45 \text{ V}$  for  $\text{NiCl}_2$  and  $\text{CuCl}_2$  electrodes,  $0$ - $0.4 \text{ V}$  for  $\text{FeCl}_3$  electrode.

Characterization. The spectral studies of aqueous electrolyte specimens were carried out at room temperature by ATR-IR technique (Thermo Nexus 6700) with an ATR cell. The internal reflection element (IRE) was diamond crystal. A Nicolet 20DXB FT-IR spectrometer was utilized to conduct the measurements of all electrolyte in the spectral range  $4000$ – $500 \text{ cm}^{-1}$ .

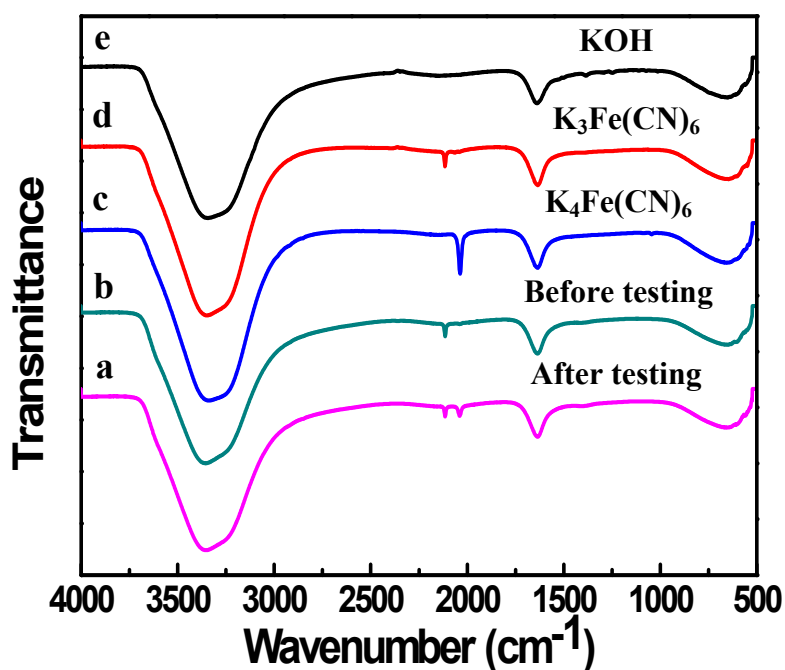


Figure S1 The global view of Fourier transform infrared spectroscopy of  $\text{K}_3\text{Fe}(\text{CN})_6$  and KOH aqueous electrolyte. Infrared spectroscopy of mixing electrolyte including 0.3 M  $\text{K}_3\text{Fe}(\text{CN})_6$  and 2 M KOH after (a) and before (b) electrochemical test and (c) 0.3 M  $\text{K}_4\text{Fe}(\text{CN})_6$ , (d) 0.3M  $\text{K}_3\text{Fe}(\text{CN})_6$ , and (e) 2M KOH aqueous solution. The infrared stretching frequency of  $\text{Fe}^{\text{III}}\text{-CN}$  was found to be  $2114\text{ cm}^{-1}$ , while the stretching frequency of  $\text{Fe}^{\text{II}}\text{-CN}$  was  $2039\text{ cm}^{-1}$ .

Table S1 Area capacitance of CuCl<sub>2</sub> electrode vs. concentration of K<sub>3</sub>Fe(CN)<sub>6</sub> and current density

Current density (mA/cm <sup>2</sup> )	Area capacitance (mF/cm <sup>2</sup> )		
	Concentration of K <sub>3</sub> Fe(CN) <sub>6</sub> (mol/L)		
	0	0.1	0.3
30	4093	7680	14133
40	3298	6160	10651
50	2456	4800	8167

Table S2 Area capacitance of NiCl<sub>2</sub> electrode vs. concentration of K<sub>3</sub>Fe(CN)<sub>6</sub> and current density

Current density (mA/cm <sup>2</sup> )	Area capacitance (mF/cm <sup>2</sup> )		
	Concentration of K <sub>3</sub> Fe(CN) <sub>6</sub> (mol/L)		
	0	0.1	0.3
30	2947	6680	9133
40	2711	6196	8018
50	2467	5689	7089

Table S3 Area capacitance of FeCl<sub>3</sub> electrode vs. concentration of K<sub>3</sub>Fe(CN)<sub>6</sub> and current density

Current density (mA/cm <sup>2</sup> )	Area capacitance (mF/cm <sup>2</sup> )		
	Concentration of K <sub>3</sub> Fe(CN) <sub>6</sub> (mol/L)		
	0	0.1	0.3
30	1410	5010	8273
40	360	2770	5590
50	71	1250	3338