Jordan flow batteries

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a, b Charge and discharge voltage profiles of various electrolytes at 400 mA g-1 along with UV-vis spectrum of ICl in these solutions recorded with time. c Cyclic voltammetry (CV) profiles of selected electrolytes with/without Cl- at 0.5 mV s-1.

Early works conducted by Kolthoff and Jordan have systematically studied the I-/I2 redox processes32,33, indicating that under specified conditions both iodide and iodine yield an anodic wave corresponding to the formation of positive iodine (I+). Further studies34,35 concluded that IO3- was the direct oxidation product of iodine in the absence of halide (Cl-, Br-) or cyanide (CN-) in an aqueous solution. This is due to the interaction between electrophilic I+ and nucleophilic species like halide36, cyanide32, and amines37 which afford to charge-transfer complexes38.

The UV-Vis absorption spectra of the less concentrated electrolytes (1-0-0, 10-0-0) resemble the tendency of I+ decomposition in the 1 m ZnSO4 solution (Fig.1b). In contrast, enhanced capability to stabilize ICl could be achieved in concentrated systems (19-19-8, 30-19-8, 30-0-0). Little intensity changes of the ICl band was observed in the whole duration without the appearance of the I2 signal.

a, b Charge and discharge voltage profiles of the 30-0-0 and 19-19-8 electrolyte from 1.25 to 1.8 V at 400 mA g-1, respectively. c, d Full-range (0.6-1.8 V) cycling performance of different systems at 400 mA g-1.

It is apparent that a suitable concentration is required to allow the normal operation of the battery to suppress the self-discharge reaction. We speculate the enhanced stability towards I+ species is correlated to the suppressed water activity and the sufficient chloride ions in the concentrated solution, which will be further discussed in the next section.

a, b Raman spectra of solutions of different concentrations. c, d Radical distribution function(RDF) of the solutions. The solid lines are the radial distribution functions, and dotted lines are the coordination numbers. e A snapshot of the MD simulation box for 19-19-8 system, along with the dominant clusters existing in such system (f). Clusters and their occurrence in the 19-19-8 system. Cyan-blue atom is Zn2+, green atom is Cl-, yellow atom is Li+, red atom is O, dark blue atom is N, brown atom is C, gray atom is Me, and white atom is H. g Summary of free water and Cl- content extracted from the MD simulation.

We conclude that in dilute ZnCl2 solutions (10-0-0, 20-0-0, and 19-5-8), high water activity rendering rapid hydrolysis of ICl, which results in low coulombic efficiency as discussed in Fig.2. Whereas in solutions with very high concentrations (30-0-0 and 30-19-8), the low Cl-activity and the substantial coordination of iodide with Zn2+ (Zn-I clusters, Supplementary Fig.5) is attributed to the rapid capacity decay. Only the 19-19-8 solution takes full advantages of low fraction of free water to suppress I+ hydrolysis and sufficient ionic conductivity with high Cl- activity to realize good performance.

a Ex situ UV vis spectrum of 19-19-8 system recorded at different charge-discharge stages. b Typical voltage-capacity profile of 19-19-8. c In situ Raman spectrum of 19-19-8 system during charge/discharge. d Diffusion coefficient versus specific capacity, calculated from GITT measurements. e CV curves with different scanning rates from 0.1 to 0.8 mV s-1. f The plots of the oxidation and reduction peak-current with the function of the root of scanning rates.

Based on the electrochemical analysis and spectral results, we depict a clear portrait of the reaction mechanism for the four-electron conversion:

The electrolyte formula (19-19-8) we proposed has the advantage of suppressed H2O activity and preserved free chloride ions, which facilitate the four-electron conversion of iodine with a high reversible capacity. While a practical battery relies on the robust anode as well, we tested the zinc anode in the 19-19-8 electrolyte to elucidate the electroplating behavior.

a, b SEM images of the Zn electrodes cycled for 1 h at 1 mA cm-2 in 1 m ZnSO4 and 19-19-8, respectively. c Coulombic efficiencies of zinc plating/stripping on Ti foil (2 mA cm-2, 1 h for the plating). d The plating/stripping in Zn||Zn symmetric cells at 1 mA cm-2 with a sweep duration of 10 min.

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