

FIG. 4 is a graph of efficiencies of one embodiment of the present invention as a function of charge-discharge cycles.

DETAILED DESCRIPTION

The following description includes the preferred best mode as well as other embodiments of the present invention. It will be clear from this description of the invention that the invention is not limited to these illustrated embodiments but that the invention also includes a variety of modifications and embodiments thereto. Therefore the present description should be seen as illustrative and not limiting. While the invention is susceptible of various modifications and alternative constructions, it should be understood, that there is no intention to limit the invention to the specific form disclosed, but, on the contrary, the invention is to cover all modifications, alternative constructions, and equivalents falling within the spirit and scope of the invention as defined in the claims.

FIGS. 1-4 show a variety of embodiments and aspects of the present invention. Referring first to FIG. 1, a diagram depicts one embodiment of a redox flow battery system. Positive electrolyte 105 and negative electrolyte 103 are delivered to an electrochemical cell 101 from storage tanks 102 and 104. The positive electrolyte comprises Fe(II) and/or Fe(III) and the negative electrolyte comprises S^{2-} and/or S. The electrolytes in the electrochemical cell are separated by a membrane or a separator 106. During discharge, the Fe(III) will be reduced to Fe(II) on the positive electrode 107, while the S^{2-} will be oxidized into elemental S on the negative electrode 108. Ions can pass through the membrane, or separator, for charge transportation and compensation. The process can be reversed during charging. Alternatively, spent electrolytes can be replaced.

FIG. 2 includes simplified redox reactions for a particular embodiment, wherein the positive electrolyte comprises 1M potassium ferricyanide [$K_3Fe(CN)_6$] and a 0.1M sodium hydroxide (NaOH) supporting solution while the negative electrolyte comprises 2M sodium sulfide in 1M NaOH supporting solution. The two flowing redox solutions were separated by a sulfonated tetrafluoroethylene based fluoropolymer-copolymer (e.g., Nafion™) membrane. The system further comprised a positive electrode (pre-treated carbon or graphite felt), and a negative electrode (Ni or Co coated carbon or graphite felt) in an electrochemical cell. Typically, during discharge, the elemental S will dissolve in the excess sodium sulfide (Na_2S) basic solution and form a sodium polysulfide (Na_2S_n) solution. The Na^+ (or some K^+ and OH^-) will pass through the Nafion™ membrane for charge transportation and compensation.

FIG. 3 includes data from the particular embodiment above showing charge-discharge cycles with the corresponding polarization curves of the positive and the negative potentials vs. Ag/AgCl (1M) reference electrodes, respectively. The absolute values of the actual cell voltage minus the potential difference between the two electrodes during charging and discharging are the ohmic losses due to the electrical resistances of the membrane and the solutions (the dish line in FIG. 3). The redox reaction between $Fe(CN)_6^{3-}$ and $Fe(CN)_6^{4-}$ was rather reversible on the positive electrode (graphite felt) during a charge-discharge cycle, while the polysulfide solution showed a higher polarization behavior during the late stages of charging. This can be attributed to the low reduction activity of element S and its concentration

polarization on the negative electrode (Co/graphite felt). The latter can be improved by modifying the negative electrode and adding appropriate amount of S into the initial Na_2S solution.

Referring to FIG. 4, the coulombic efficiency of the same embodiment was over 93% and the energy efficiency was over 75%. These values are comparable to the state-of-art bromide-polysulfide flow battery system.

While a number of embodiments of the present invention have been shown and described, it will be apparent to those skilled in the art that many changes and modifications may be made without departing from the invention in its broader aspects. The appended claims, therefore, are intended to cover all such changes and modifications as they fall within the true spirit and scope of the invention.

We claim:

1. A redox flow battery system comprising a positive electrolyte comprising Fe(III) and Fe(II) in a positive electrolyte supporting solution, a negative electrolyte comprising S^{2-} and S in a negative electrolyte supporting solution, and a membrane or a separator contacting and separating the positive electrolyte solution and the negative electrolyte solution, the positive electrolyte and negative electrolyte supporting solutions both having pH values greater than or equal to 6.

2. The system of claim 1, wherein concentrations of the Fe(II) and Fe(III) are greater than 0.2 M.

3. The system of claim 1, wherein concentrations of the S^{2-} and S are greater than 0.1 M.

4. The system of claim 1, having a cell temperature between $-10^\circ C.$ and $60^\circ C.$ during operation.

5. The system of claim 1, wherein the negative electrolyte comprises a polysulfide compound.

6. The system of claim 1, having a state-of-charge condition greater than 0% and less than 100% during operation.

7. The system of claim 1, wherein the system further comprises electrodes, wherein both electrodes comprise carbon.

8. The system of claim 7, wherein the carbon electrodes comprise graphite felt.

9. The system of claim 7, wherein the carbon electrodes further comprise a Ni coating or a Co coating.

10. The system of claim 1, wherein the system further comprises electrodes, wherein both the positive electrode and the negative electrode comprise noble metals.

11. The system of claim 10, wherein the noble metal electrodes comprise a metal selected from the group consisting of Ni, Co, Au, Pt, and stainless steels.

12. The system of claim 1, wherein the membrane or the separator comprises a sulfonated tetrafluoroethylene based fluoropolymer-copolymer.

13. A redox flow battery system comprising a positive electrolyte comprising Fe(III) and Fe(II) in a positive electrolyte supporting solution, a negative electrolyte comprising S^{2-} and S in a negative electrolyte supporting solution, and a membrane or a separator contacting and separating the positive electrolyte solution and the negative electrolyte solution, the positive electrolyte and negative electrolyte supporting solutions comprising alkali metal hydroxide, ammonium hydroxide, or both.

14. The redox flow battery system of claim 13, wherein the positive electrolyte and negative electrolyte supporting solutions both have values greater than or equal to 6.

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