



# High performance of zinc-ferrum redox flow battery with $\text{Ac}^-/\text{HAc}$ buffer solution

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## ABSTRACT

A green low-cost redox flow battery using Zn/Zn<sup>2+</sup> redox couple in HAc/NaAc medium and Fe<sup>2+</sup>/Fe<sup>3+</sup> redox couple in H<sub>2</sub>SO<sub>4</sub> medium was first proposed and investigated for potential stationary energy storage applications. The presence of HAc/NaAc in the negative electrolyte can keep the pH between 2.0 and 6.0 even when a large amount of H<sup>+</sup> ions move into negative electrolyte from positive electrolyte through ion exchange membrane. In the pH range of 2.0–6.0, the chemical reaction of Zn species with H<sup>+</sup> species is very insignificant; furthermore, the electroreduction of H<sup>+</sup> ion on the negative electrode is significantly suppressed at this pH range. The zinc-ferrum redox flow battery (Zn/Fe RFB) operated within a voltage window of 0.5–2.0 V with a nearly 90% utilization ratio, and its energy efficiency is around 71.1% at room temperature. These results show that Zn/Fe RFB is a promising option as a stationary energy storage equipment.

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## 1. Introduction

Renewable energy technologies have attracted considerable scientific and social attention in recent years. Solar and wind are probably the most abundant and readily accessible among the available renewable energy sources, and are increasingly being recognized as important components of future global energy production [1]. However, it makes their use and dispatch through the aging grid quite challenging because of the changeable nature of the power from these intermittent renewable sources. One effective way to smooth out the intermittency is to employ electrical energy storage. Redox flow batteries (RFBs) are capable of storing large amount of power or energy in a pair of oxidized and reduced species dissolving in two separate liquid electrolytes [2,3]. The conversion between the electrical energy and chemical energy takes place when the electrolytes flow through electrodes separated by an ionic conducting membrane. Storage of energy in liquid electrolytes instead of in electrodes makes RFB cycle without suffering mechanical or structural stress in their electrodes, which is superior over the conventional batteries such as Li-ion battery [4,5]. The power of the RFB system is determined by the size of the electrode whereas the energy storage capacity is determined by the volume and concentration of the electrolyte.

Fe/Cr redox flow battery invented by Thaller in 1975 used the Fe<sup>2+</sup>/Fe<sup>3+</sup> halide solution as positive electrolyte and the Cr<sup>2+</sup>/Cr<sup>3+</sup> halide solution as negative electrolyte, which suffered from cross-contamination of electrolytes [6]. All-vanadium redox flow battery developed in the 1980s overcomes the cross-contamination issue using the same element vanadium in both negative electrolyte and positive electrolyte [7–9]. In addition, a number of other redox flow batteries were reported, including V/Br RFB [10], Zn/Br RFB [11], and Zn/Ce RFB [12–16].

While great progress was made in advancing RFBs, challenges remain to completely meet the environment-requirement and cost-requirement for broad application. For example, Zn/Br RFB system demonstrates an excellent electrochemical reversibility, but the technology is significantly limited by the strong corrosivity of Br<sub>2</sub>. As for Fe/Cr RFB system, the inferior activity of the Cr<sup>2+</sup>/Cr<sup>3+</sup> electrochemical reaction requires a combination of a catalyst-loaded electrode and elevated operation temperature to enhance its redox reaction. The strong-oxidation ability of Ce<sup>4+</sup> species in positive electrolyte of Zn/Ce RFB limits the material choices for a membrane. Although significant progress has been made in free-membrane Zn/Ce RFB [17], the high oxidation reactivity of Ce<sup>4+</sup> still remains a concern for electrode corrosion.

Gong et al. reported a zinc-iron redox flow battery [18] based on double-membrane triple-electrolyte design. ViZn Energy has been working on zinc/ferro-ferricyanide redox flow system [19] for several years, and this system is demonstrating over 10,000 cycles with a circulating aqueous alkaline electrolyte to overcome

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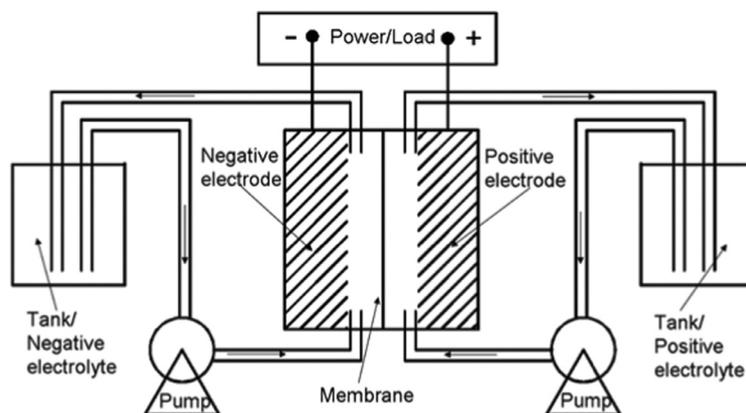
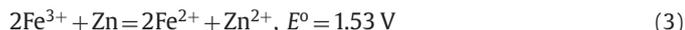


Fig. 1. Schematic diagram of the Zn/Fe RFB.

the problems of hydrogen evolution and corrosion, a zinc negative electrode which takes advantage of the high energy capacity of zinc, and an inert cathode. However, the toxic properties of ferro-ferricyanide limit its application. Different from the zinc/ferro-ferricyanide RFB, Zn/Fe RFB proposed by us in this paper is a green system that employs Zn/Zn<sup>2+</sup> as the negative redox couple and Fe<sup>2+</sup>/Fe<sup>3+</sup> as the positive redox couple. Particularly, HAc/NaAc buffer solution is used as negative electrolyte supporting medium and H<sub>2</sub>SO<sub>4</sub> as positive electrolyte supporting medium. A standard voltage of 1.53 V can be obtained based on the following redox reaction proceeding at the negative and positive electrode respectively:



The electrochemical performance of a Zn/Fe system has demonstrated a successful capitalization on the combined benefits of the Zn/Ce and Fe/Cr flow battery systems while circumventing their intrinsic issues. Compared with the Cr<sup>2+</sup>/Cr<sup>3+</sup> couple, the Zn/Zn<sup>2+</sup> couple possesses a much better electrochemical activity, which would free the system from using the catalyst and the high-temperature management system. The lower potential of the Fe<sup>2+</sup>/Fe<sup>3+</sup> couple at the positive electrode of the Zn/Fe cell also reduces its corrosive strength compared with the Zn/Ce RFB, allowing lower-cost membranes to be potentially employed with resultant cost reduction. The production cost for the Zn/Fe RFB system equates to around 80% of Zn/Ce RFB system. Hence, this system has tremendous application futures in the development of new energy.

The purpose of this study centered on experimentally investigating the performance of Zn/Fe RFB that employs the Zn/Zn<sup>2+</sup> redox couple at the negative electrode and the redox couple Fe<sup>2+</sup>/Fe<sup>3+</sup> at the positive electrode. This paper reports the details of the work.

## 2. Experimental

### 2.1. Chemicals

Analytical grade zinc carbonate basic (Aladdin, 3Zn(OH)<sub>2</sub>•2ZnCO<sub>3</sub>), sodium acetate anhydrous (Aladdin, CH<sub>3</sub>COONa, NaAc), acetate (Aladdin, CH<sub>3</sub>COOH, HAc), concentrated sulfuric acid (Zhejiang Zhuguang Group Sanying Colored Lantern Co. Ltd., H<sub>2</sub>SO<sub>4</sub>, 98%), concentrated hydrochloric acid (Shenzhen Dichuang Chemical Co. Ltd., HCl, 38%), iron (II) chloride tetrahydrate (Aladdin, FeCl<sub>2</sub>•4H<sub>2</sub>O, > 99.0%), were used as

received. Zinc foil and carbon felt were purchased from Shanghai Yitian Metalwork Co. Ltd., and Xi'an Carbon Materials Co. Ltd., respectively. HZ115 ion exchange membrane (Baoying Runhua Electrostatic Coating Engineering Co. Ltd., China) was used as the separator for Zn/Fe RFB. All solutions were prepared with doubly distilled water. Zinc sulfate was prepared by stirring zinc carbonate basic in 5 mol/L sulfuric acid, using a PTFE-coated steel magnetic stirred bar (Beijing Ruicheng Albert Instrument Equipment Co. Ltd., 1 cm diameter, 4 cm length). The resulting zinc sulfate solution was colorless with no precipitate.

### 2.2. Cyclic voltammetry

A typical three-electrode glass cell with approximately 50 mL of volume was used for the cyclic voltammetry (CV). This glass cell was equipped with a water jacket connected to a water thermostat controller (DC-0506, Shanghai Qiqian Electronic Technology Co. Ltd.) to maintain the temperature of 25°C. For the half-cell reaction of Zn/Zn<sup>2+</sup>, the working electrodes (area: 0.36 cm<sup>2</sup>) were graphite and glassy carbon. The counter electrode was a platinum mesh (area: 5 cm<sup>2</sup>). The electrode potentials were measured against a saturated Ag/AgCl electrode. The electrochemical measurements were made using an electrochemical station (CHI660, USA).

### 2.3. Flow battery experiments

The performance of Zn/Fe RFB was evaluated under a constant current using a laboratory-scale cell that was connected with an eight-channel battery test system (BTS-5V3A, Neware Ltd., China), as shown in Fig. 1. The overall volume of the cell is 4.43 cm × 4.43 cm × 4.30 cm, divided into two equal parts of 4.43 cm × 4.43 cm × 2.15 cm by an ion exchange membrane (HZ115, China). The electrolytes were stored in two external tanks (each 100 mL), pumped at a flow rate of 12.6 mL/min (Peristaltic pump BT100-1L) through the electrodes where the electrochemical reactions occurred. The electrodes were immersed in solution approximately 4.0 cm. Zinc foil and carbon felt were selected as electrode materials for Zn/Fe RFB. In one case, zinc foil (6 cm × 4 cm × 0.2 cm) and carbon felt (6 cm × 4 cm × 1 cm) were employed as negative electrode and positive electrode, respectively; in another case, carbon felts were used as both negative electrode and positive electrode. Carbon felt was thermally treated in air at 450 °C for 3 h with the aim of improving its hydrophilicity. Iron (II) chloride aqueous solution of 1 mol/L with 1.5 mol/L H<sub>2</sub>SO<sub>4</sub> was used as positive electrolyte. In one case, negative electrolyte consisted of 1 mol/L ZnSO<sub>4</sub>, 1.5 mol/L HAc, and 1.5 mol/L NaAc; in another case, it only was zinc sulfate aqueous solution of 1 mol/L.

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