



A novel iron-lead redox flow battery for large-scale energy storage



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HIGHLIGHTS

- A redox flow battery using low-cost iron and lead redox materials is presented.
- Fe(II)/Fe(III) and Pb/Pb(II) redox couples exhibit fast kinetics in the MSA.
- The energy efficiency of the battery is as high as 86.2% at 40 mA cm⁻².

ARTICLE INFO

Article history:

Received 9 November 2016
Received in revised form
7 January 2017
Accepted 8 February 2017

Keywords:

Flow battery
Iron-lead redox flow battery
Energy storage
Energy efficiency
Cycle stability

ABSTRACT

The redox flow battery (RFB) is one of the most promising large-scale energy storage technologies for the massive utilization of intermittent renewables especially wind and solar energy. This work presents a novel redox flow battery that utilizes inexpensive and abundant Fe(II)/Fe(III) and Pb/Pb(II) redox couples as redox materials. Experimental results show that both the Fe(II)/Fe(III) and Pb/Pb(II) redox couples have fast electrochemical kinetics in methanesulfonic acid, and that the coulombic efficiency and energy efficiency of the battery are, respectively, as high as 96.2% and 86.2% at 40 mA cm⁻². Furthermore, the battery exhibits stable performance in terms of efficiencies and discharge capacities during the cycle test. The inexpensive redox materials, fast electrochemical kinetics and stable cycle performance make the present battery a promising candidate for large-scale energy storage applications.

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1. Introduction

The massive utilization of intermittent renewable energy sources especially wind and solar energy urgently needs large-scale energy storage devices to achieve uninterrupted and reliable power supply [1–4]. The redox flow battery (RFB) is one of the most promising large-scale energy storage solutions due to its striking features including easy scalability, high energy efficiency, intrinsic safety and long lifetime [5]. Over the past decades, various RFB systems including all-vanadium (all-V), all-copper (all-Cu), soluble all-lead (all-Pb), iron-chromium (Fe/Cr), iron-vanadium (Fe/V) and zinc-cerium (Zn/Ce) RFBs have been developed [6–17].

One of the most significant challenges for RFBs is to develop the cost-effective and stable redox material with good electrochemical kinetics, suitable redox potential and negligible side reactions. The vanadium redox flow battery (VRFB) has been widely studied, and demonstrates excellent battery performance [18–24]. However, its

commercialization is hindered by the high costs of the vanadium material and perfluorosulfonic acid (PFSA) membrane. The iron-chromium redox flow battery (ICRFB) utilizes inexpensive iron and chromium redox materials, and has achieved a high output power density in the recent studies [25,26]. However, the low redox potential of the Cr(II)/Cr(III) redox couple (−0.41 V vs SHE) causes the hydrogen evolution issue, which induces technical challenges for the long-term operation of the ICRFB [27]. The other RFBs using low-cost redox materials, such as all-Cu, all-Fe and all-Pb RFBs, have also been studied, but their performances are relatively low and need to be further improved [6,12,28–30]. The energy efficiency of the all-Cu RFB is merely 50% at 20 mA cm⁻² (40 °C) due to the slow kinetics of Cu/Cu(I) redox couple [31]. Lloyd et al. reported that an all-Cu RFB delivers an output voltage of 0.55 V at 80 mA cm⁻² (60 °C) [32]. In the all-Fe RFB, the low redox potential of the Fe/Fe(II) redox couple (−0.44 V vs SHE) causes hydrogen evolution and coulombic loss. The sluggish kinetics of the Fe/Fe(II) redox couple deteriorates the performance of the all-Fe RFB [30]. The reversibility and kinetics of the Pb(II)/Pb(IV) redox couple are poor, which limits the all-Pb RFB to a low operating current density of 20 mA cm⁻² with the energy efficiency of 79% [33]. The

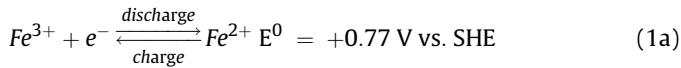
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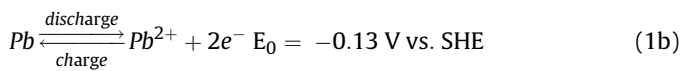
computationally assisted strategies have also been developed to discover and optimize the new electrolyte composition for electrochemical energy storage devices [34–36].

In this work, a novel iron-lead redox flow battery (Fe/Pb RFB) is demonstrated. As illustrated in Fig. 1, the Fe/Pb RFB utilizes the low-cost Fe(II)/Fe(III) and Pb/Pb(II) redox couples in the methanesulfonic acid (MSA) as the positive and negative redox materials, respectively, which are separated by an ion-exchange membrane or a separator. The working principle that the Fe/Pb RFB stores and releases electricity can be expressed as follows:

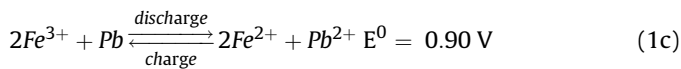
Positive electrode:



Negative electrode:



Overall reaction:



During the charge process, the Fe(II) ions at the positive electrode release the electrons, and oxidize to Fe(III) ions; the Pb(II) ions at the negative electrode acquire the electrons from the external circuit, and electrodeposit onto the electrode as the metallic Pb. During the discharge, the electrochemical process is reversed.

At the positive electrode, the Fe(II)/Fe(III) redox couple generally exhibits fast kinetics and high solubility in the strong acids [7]. At the negative electrode, the Pb/Pb(II) redox couple achieves high solubility and facile kinetics in the MSA, which has been studied in the soluble all-Pb RFBs [14,28,29]. Metallic lead has a high overpotential to mitigate hydrogen evolution, and allows the use of highly conductive MSA [33], which has been widely studied as supporting electrolyte in all-V, all-Pb and Zn-Ce RFBs [1]. To address the cross-contamination issue when composing the present RFB using different elements as positive and negative redox materials, the identical mixed-reactant solution containing premixed iron and lead salts is proposed for both the initial positive and negative electrolytes of 0% state-of-charge (SOC) in the present RFB. With the mixed-reactant solution, the capacity loss caused by species

crossover can be simply recovered by re-mixing and re-separating the positive and negative electrolytes [13,37].

2. Experimental

2.1. Electrolyte preparation

The premixed iron and lead electrolyte was prepared by dissolving the lead monoxide (Nacalai Tesque) and iron powder into the diluted methanesulfonic acid (Sigma-Aldrich). During the preparation process, the nitrogen purging was used to protect the Fe(II) from being oxidized by the air.

2.2. Cyclic voltammetry

Cyclic voltammetry (CV) experiments were conducted using a three-electrode system on a workstation (Autolab, PGSTAT30). A glassy carbon electrode (GCE), platinum mesh electrode and saturated calomel electrode (SCE) served as the working electrode, counter electrode and reference electrode, respectively. The 100 mL of mixed-reactant solution (0.1 M $Fe(SO_3CH_3)_2$ + 0.1 M $Pb(SO_3CH_3)_2$ + 2.0 M HSO_3CH_3) was used for the CV test. The scan rates were 20, 30, 40, 50 and 100 $mV s^{-1}$. The measurement was performed at room temperature (25 °C).

2.3. Battery test

The carbon cloth (ELAT-H, 0.4 mm) was pretreated in air at 600 °C for 5 h, and served as the electrodes. For the negative side, the flow cavity was machined on the graphite plate. One layer of carbon cloth with an active area of 2.0 cm × 2.0 cm was placed near the wall of the cavity, and served as the negative electrode. A 3.0-mm-thick non-conductive polyacrylonitrile porous felt contacted the carbon cloth, and served as the cavity filler. The Nafion® NR-212 membrane was employed without pretreatment. For the positive side, an interdigitated flow field with the channel depth 1.5 mm, the channel width 1.0 mm and rib width 1.0 mm was grooved on the graphite plate. The two layers of carbon cloth served as the positive electrode. The polytetrafluoroethylene gaskets were used for sealing. The gold-coated copper plates were used as the current collectors. 40 mL identical mixed-reactant solutions (0.6 M $Fe(SO_3CH_3)_2$ + 0.3 M $Pb(SO_3CH_3)_2$ + 2.0 M HSO_3CH_3) were used as the positive and negative electrolyte, respectively, for the charge-discharge test. The electrolytes were circulated in the Norprene® #16 Chemical Tubing by a 2-channel peristaltic pump (Longer pump, WT600-2J). The charge-discharge tests were conducted on Arbin BT2000 at current densities of 40, 80 and 120 $mA cm^{-2}$ with a voltage window of 0.4–1.1 V. The flow rate of the electrolyte was 50 $ml min^{-1}$. The internal resistance of the battery was measured by the built-in function of Arbin BT2000 using a current interrupt method [38]. The cycle test was conducted at 80 $mA cm^{-2}$ with 25 mL identical solutions (0.5 M $Fe(SO_3CH_3)_2$ + 0.25 M $Pb(SO_3CH_3)_2$ + 2.0 M HSO_3CH_3) as the positive and negative electrolyte, respectively. The coulombic efficiency (CE), voltage efficiency (VE) and energy efficiency (EE) of the battery are calculated as the following equations:

$$CE = \frac{t_d}{t_c} \times 100\% \quad (2a)$$

$$VE = \frac{V_d}{V_c} \times 100\% \quad (2b)$$

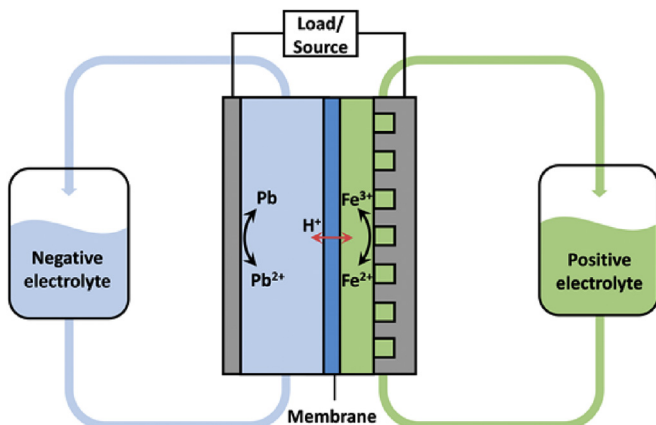


Fig. 1. Schematic of an iron-lead redox flow battery.

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