



A high power density single flow zinc–nickel battery with three-dimensional porous negative electrode

Yuanhui Cheng^{a,b}, Huamin Zhang^{a,**}, Qinzhi Lai^{a,*}, Xianfeng Li^a, Dingqin Shi^a, Liqun Zhang^{a,b}

^aDivision of Energy Storage, Dalian Institute of Chemical Physics, Chinese Academy of Sciences, 457 Zhongshan Road, Dalian 116023, PR China

^bUniversity of Chinese Academy of Sciences, Beijing 100039, PR China

HIGHLIGHTS

- Three-dimensional porous electrode is first introduced to ZNBs.
- The utilized electrode area and potential distribution has been studied.
- An EE (80.1%) is obtained at 80 mA cm⁻², which is the highest value ever reported.
- The power density is improved nearly four times to 83 W Kg⁻¹.

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ABSTRACT

Low power density (operated current density) is one critical obstacle to the development of single flow zinc–nickel batteries (ZNBs). Three-dimensional porous nickel foams (NFs) are first introduced to ZNBs to improve power density. The relationship between utilized electrode area, potential distribution and operated current density has been studied. A high coulombic efficiency (97.3%) and energy efficiency (80.1%) are obtained at 80 mA cm⁻² over 200 cycles, which is the highest value ever reported for ZNBs. The power density is improved nearly fourfold to 83 W kg⁻¹. The results indicate that three-dimensional porous electrodes are more suitable as negative electrodes for ZNBs under high operated current densities. This provides an effective way to improve power density of ZNBs and will arouse a new revolution in the development of flow batteries.

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1. Introduction

Exigent demand for energy has attracted the attention on the applications of clean energy resources. But these clean energy forms (e.g., solar, wind, nuclear and tide) are usually fluctuant and dispersed, which make them difficult to integrate into the electric grid or utilize continuously. Hence, energy storage becomes a critical technology for the applications of clean energy resources [1,2]. Energy storage mainly focuses on electrochemical systems due to its wide range of storage capacity and no regional condition confine [3,4]. Among them, flow battery (FB) is one of the most promising candidates due to its high storage efficiency, excellent capability, security, reliability and flexible operation [5,6]. Thus, various flow battery systems have sprung up attributed to the

interest in both researches and commercial applications [7–15]. One of the most promising systems, zinc–nickel batteries are of high open circle potential (1.705 V) and high energy density (above 65 Wh kg⁻¹) [15]. Furthermore, ZNBs are made from cheap, non-toxic materials that are inherently safe. No membrane is needed, which simplifies the battery system and reduces the integration cost. But together with other demonstrated systems, they are nearly all operated at current densities below 40 mA cm⁻². For instance, the operated current density of soluble lead acid batteries (SLABs) [14], zinc-bromine batteries (ZBBs) [9,16] and zinc–nickel batteries (ZNBs) [17,18] is below 20 mA cm⁻². The polysulfide–bromine battery (PSB) reaches to 40 mA cm⁻² [13], but still limits the quick response for energy conversion. The power density, which is associated with the operated current density, has a significant influence on energy conversion rate and the integration cost. Thus, increasing power density of ZNBs without compromising battery performance is an effective strategy to meet the requirements for commercialization.

* Corresponding author. Tel./fax: +86 (0) 411 84379580.

** Corresponding author.

E-mail address: qinzhilai@dicp.ac.cn (Q. Lai).

In ZNBs, both anolytes and catholytes are the high soluble alkaline zincate solutions pumped through a single pump. During charge, metallic zinc deposits at the negative electrode at -1.216 V vs. the normal hydrogen electrode (NHE), whereas $\text{Ni}(\text{OH})_2$ undergoes solid-phase transformation to NiOOH at positive electrode at 0.49 V vs. NHE. For alkaline batteries, spongy zinc formation upon charging is a critical issue, which limits the lifetime. Many studies have been focused on the deposited morphology of zinc electrodes. Wen et al. [18] have studied lead ion and tetrabutylammonium bromide as additives for zinc deposition. But these additives usually increase the overpotential for zinc deposition and reduce the storage efficiency. Further, the hydrogen evolution is another issue especially under high current densities [19]. This evolved gas reduces energy efficiency and makes zinc particles peeling off without reaction. That's why ZNBs are still operated at low current densities (below 20 mA cm^{-2}), which limits the power density and further development.

However, no study has successfully solved the two issues without compromising battery performances and environmental concerns. Three-dimensional porous nickel foams (NFs) have been widely used in batteries, electrochemical supercapacitors and fuel cells as the current collector and support matrix with open-pore structures and high specific surface areas [20–22]. The open-pore structure of three-dimensional electrode considerably reduces the internal resistance of the interface between electrode and electrolyte, and prevents zinc from falling before reacted. Their high specific surface is generally considered to reduce polarization with low realistic current density per geometrical area [23]. Therefore, three-dimensional NFs may be more suitable to act as negative electrodes for ZNBs, especially under high operated current densities. Whereas, according to the previous studies [24,25], as the three-dimensional porous electrodes usually have a considerable thickness, the potential distribution is non-uniform resulting in low utilization, it is of great importance to investigate the relationship between utilized electrode area, potential distribution and operated current density combined with battery performances.

In this work, we aimed at finding a more suitable negative electrode to improve the power density of ZNBs. Two types of nickel electrodes were tested to explore the relationship between electrode structure and battery performance. Furthermore, the relationship between utilized electrode area, potential distribution and operated current density was studied.

2. Experimental section

2.1. Electrochemical measurement

The electrochemical experiments were carried out in a conventional three-electrode cell using a CHI 612C potentiostat (Shanghai Chenhua Instrumental Co., Ltd., China). The working electrode was a piece of NF ($3 \times 5 \times 2$ mm, 110 pores per inch, 420 g m^{-2} , Changsha Lyrun Material Co., Ltd., China) or a piece of nickel sheet (NS) ($3 \times 5 \times 0.1$ mm, 890 g m^{-2} , Shanghai Jinchang Alloy Co., Ltd., China) with a sintered nickel hydroxide ($10 \times 10 \times 0.7$ mm, Jiangsu Highstar Battery Manufacturing, China) acting as the counter electrode. The reference electrode was an Hg/HgO electrode, which has a potential of 0.98 V vs. NHE. The NF and NS were degreased with acetone, etched with 6.0 mol dm^{-3} HCl for 10 min and rinsed thoroughly with deionized water. The electrolyte was 8.0 mol dm^{-3} KOH containing 0.4 mol dm^{-3} ZnO. All solutions were made with analytical grade chemical reagents and deionized water. The solutions were purged by bubbling ultra purity nitrogen stream for 30 min prior to measurements and maintained with a slight overpressure of nitrogen during the experiments. All

potentials were referred to the reference electrode. Current densities were normalized to the geometrical area of working electrodes.

2.2. ZNB single cell test

The charge–discharge tests were conducted in a two-electrode cell constituted by a sintered nickel hydroxide positive plate ($30 \times 30 \times 0.7$ mm, area capacity, 25 mAh cm^{-2}) and a negative plate NF ($30 \times 30 \times 2$ mm) or NS ($30 \times 30 \times 0.1$ mm). The electrodes were pre-treated before test. The schematic diagram of cell structure was shown in Fig. 1. The interelectrode gap was 5 mm. The electrolyte was circulated through the cell and reservoir by a pump. Constant current charge–discharge tests were conducted by Arbin BT-2000 (Arbin Instruments, America). In order to avoid excessive gas evolution and degradation of electrode materials, the cell was charged to 80% of its full capacity based on the sintered nickel hydroxide electrode, and discharged at the same current density to 0.8 V. All above experiments were performed at room temperature.

2.3. Overpotential measurement

An Hg/HgO electrode was settled between positive and negative electrodes to monitor the variety of potentials on them using another two channels of Arbin BT-2000. The values were recorded every 5 s during the batteries' charge–discharge processes.

2.4. SEM characterization

The surface morphologies of deposited zinc were detected by scanning electron microscopy (SEM, JEOL JSM-6000, JEOL Ltd., Japan). Images were acquired using a 15 kV accelerating voltage. Negative electrodes after charging process were washed with deionized water and dry in a vacuum desiccator before test.

3. Results and discussion

3.1. Electrochemical performance

Electrochemical measurements are conducted to investigate zinc deposition/dissolution and hydrogen evolution on different

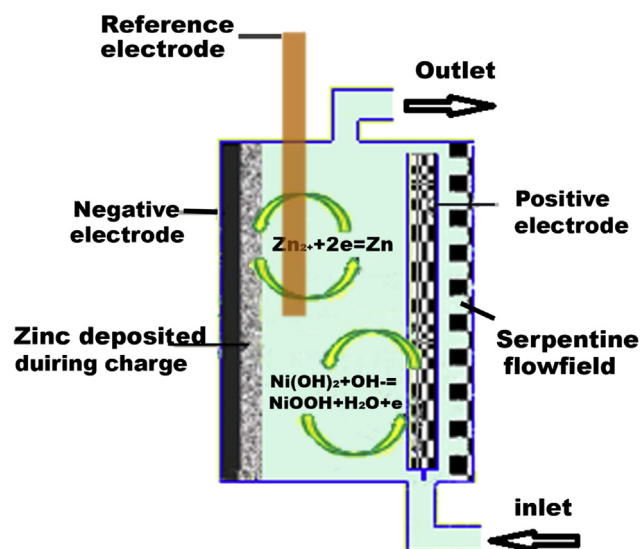


Fig. 1. Schematic diagram of cell structure.

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