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Flow distribution and maximum current density studies in redox flow batteries with a single passage of the serpentine flow channel



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HIGHLIGHTS

- A mathematical model of a fuel cell-like flow battery has been developed.
- The model predicts a small flow penetration into a carbon paper electrode.
- Reactant penetrating an electrode limits the current density of the flow battery.
- Predicted maximum current densities match published experimental results.

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ABSTRACT

Flow batteries show promise for very large-scale stationary energy storage such as needed for the grid and renewable energy implementation. In recent years, researchers and developers of redox flow batteries (RFBs) have found that electrode and flow field designs of PEM fuel cell (PEMFC) technology can increase the power density and consequently push down the cost of flow battery stacks. In this paper we present a macroscopic model of a typical PEMFC-like RFB electrode-flow field design. The model is a layered system comprised of a single passage of a serpentine flow channel and a parallel underlying porous electrode (or porous layer). The effects of the inlet volumetric flow rate, permeability of the porous layer, thickness of the porous layer and thickness of the flow channel on the flow penetration into the porous layer are investigated. The maximum current density corresponding to stoichiometry is estimated to be 377 mA cm⁻² and 724 mA cm⁻², which compares favorably with experiments of ~400 mA cm⁻² and ~750 mA cm⁻², for a single layer and three layers of the carbon fiber paper, respectively.

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1. Introduction

Redox flow batteries have gained unprecedented attention for energy storage applications. Well-suited for and scalable to utility scale storages, they can be employed to cope with intermittent power generation from renewable energy sources, such as solar and wind energy and are a useful tool for load leveling, peak shaving and emergency back up to improve the stability of the power grid [1,2]. Several research groups are currently

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working on redox flow batteries. Skyllas-Kazacos et al. [3,4] developed an all-vanadium redox flow battery (all-VRFB) that was motivated by earlier studies on iron-chrome as a redox couple [5]. An advantage of the all-VRFB is that species crossover leads to only a temporary capacity loss in contrast to a permanent capacity loss for the flow cells with two or more different elements acting as anions or cations in the electrolyte; examples being iron-chromium [6], bromine-polysulfide [7] and zinc-bromine [8], etc. Most recent studies are focused on vanadium flow cells, especially for the all-VRFB. Savinell et al. [9] developed an all-iron flow cell with a potential advantage of very low costs. Chiang et al. [10] invented a semi-solid lithium rechargeable flow battery within using active slurry porous electrode material through reaction chambers and an improved current density is achieved. Aziz et al. [11] has introduced an organic & inorganic



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RFB with quinone reactants in an aqueous electrolyte. High current density and discharge capacity retention have been demonstrated.

Computational modeling approaches have been reported to explore performance improvements in RFB flow cells [12–18]. Walsh et al. developed several mathematical flow battery models [14–17] that describe isothermal [14], non-isothermal [15], hydrogen [16] and oxygen evolution [17] (with bubble formation) transport processes in electrochemical reactions. These models include diffusion, convection and migration. A dilute-solution approximation method was incorporated. Vynnycky [18] proposed an asymptotic model for a large-scale cell stack to save computational time while improving the calculation efficiency.

Few experimental and numerical studies on details of flow fields for RFBs have been reported. Kjeang et al. [19] designed a microfluidic vanadium redox flow cell with a high aspect ratio Yshape micro flow channel over a porous carbon substrate to obtain a peak power density of 70 mW cm⁻², which is small for large-scale energy storage applications. Recently, Zawodzinski et al. [20–22] and Mench et al. [21,22] designed flow cell stack configurations for RFBs to improve current density and power density. These designs are based on modern cell designs employed in PEM fuel cells and allow operation at much higher current densities as compared to earlier conventional flow batteries. In these designs the electrolyte flows through serpentine channels behind a porous electrode. This is different from the conventional flow battery designs where the electrolyte flows through a carbon felt electrode. Specifically, a polarization curve analysis method [20] was used to analyze performance losses in two types of flow cell configurations: (a) a simple battery (SB) with a carbon felt electrode embedded in the PVC compartments and (b) a modified fuel cell with a serpentine flow channel fuel cell battery (FCB) over the carbon felt (FCB-F) or the carbon fiber paper electrode (FCB-P). A higher limiting current density was obtained in the FCB-P than in the SB. A peak power density of 557 mW cm⁻², which is more than five times that obtained using conventional flow cell configurations, was achieved at 60% state of charge by adopting a "zero-gap" serpentine flow channel flow battery architecture [21]. A superior behavior for lower over-potential was observed in the flow cells composed of two negative porous electrode layers. The potential effects of thermally pretreated porous electrodes and ion selective membranes on the performance of the "zero-gap" serpentine flow cell were also investigated [22]. A peak power density of 767 mW cm⁻² was obtained by using three layers of carbon fiber paper with an oxidative thermal pretreatment and the Nafion 212 membrane. It was pointed out that surface morphologies and oxygen concentrations might alter the surface functionalization.

Chen et al. [23] conducted experiments on a parallel flow structure together with CFD analysis for flow distributions in the all-VRFB, but generated a very low power density of 15.9 mW cm^{-2} . It appears that flow rates may be highly variable



Fig. 1. The flow cell compartment: 1-end plates; 2-current collectors; 3-graphite plates with serpentine flow channels; 4-gaskets; 5-porous electrodes; 6-ion selective membrane; 7-load; 8-electrolyte tanks and 9-pumps.

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