



# Performance enhancement of iron–chromium redox flow batteries by employing interdigitated flow fields



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## HIGHLIGHTS

- The ICRFBs with the interdigitated and serpentine flow fields are investigated.
- The IFF design enhances species transport in the porous electrode.
- The IFF design enables more uniform catalyst distribution in the porous electrode.
- The energy efficiency of the ICRFB with the IFF reaches 80.7% at 320 mA cm<sup>-2</sup>.

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## ABSTRACT

The catalyst for the negative electrode of iron–chromium redox flow batteries (ICRFBs) is commonly prepared by adding a small amount of Bi<sup>3+</sup> ions in the electrolyte and synchronously electrodepositing metallic particles onto the electrode surface at the beginning of charge process. Achieving a uniform catalyst distribution in the porous electrode, which is closely related to the flow field design, is critically important to improve the ICRFB performance. In this work, the effects of flow field designs on catalyst electrodeposition and battery performance are investigated. It is found that compared to the serpentine flow field (SFF) design, the interdigitated flow field (IFF) forces the electrolyte through the porous electrode between the neighboring channels and enhances species transport during the processes of both the catalyst electrodeposition and iron/chromium redox reactions, thus enabling a more uniform catalyst distribution and higher mass transport limitation. It is further demonstrated that the energy efficiency of the ICRFB with the IFF reaches 80.7% at a high current density (320 mA cm<sup>-2</sup>), which is 8.2% higher than that of the ICRFB with the SFF. With such a high performance and intrinsically low-cost active materials, the ICRFB with the IFF offers a great promise for large-scale energy storage.

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## 1. Introduction

Deployment of intermittent renewable energy sources such as wind and solar energy has been increasing substantially, which raises an urgent demand to develop the large-scale energy storage devices for continuous and reliable power output [1–3]. The redox flow battery (RFB) has attracted extensive interests as a promising large-scale energy storage technology due to its unique advantages including ease of scalability, long cycle life, intrinsic safety and high efficiency [3]. In the past decades, various RFB systems have been proposed and developed [3–16]. However, the current RFB technologies still have not met the stringent cost and performance

requirements for the broad penetration of energy storage market.

The iron–chromium redox flow battery (ICRFB) utilizes the low-cost and benign Fe(II)/Fe(III) and Cr(II)/Cr(III) redox couples in the acid supporting medium as the catholyte and anolyte, respectively [2,17]. The cost of chromium and iron active materials used in ICRFBs is estimated to be as low as \$17 kWh<sup>-1</sup>, which provides the ICRFB a sufficient basis and great possibility to be a cost-effective energy storage system [2,17,18].

Historically, the ICRFB adopts a flow-through cell structure, in which the electrolyte is directly pumped through the porous electrode [19]. To bypass high pump loss, the conventional ICRFBs have thick electrodes (generally 3.0–6.0 mm) [20–22], leading to a high ohmic resistance [23,24]. For this reason, the conventional ICRFB is limited to a low operating current density of 80 mA cm<sup>-2</sup>, resulting in bulky and costly cell stacks [18,25]. Recently, an ICRFB

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with the serpentine flow field (SFF) cell structure and carbon paper electrodes enables a high operating current density of  $200 \text{ mA cm}^{-2}$ , primarily due to a reduced ohmic loss [26].

In addition to the ohmic loss, the activation loss plays an important role in the battery performance, which is closely associated with the active surface area and electrochemical kinetics of redox couples. The Fe(II)/Fe(III) redox couple has an excellent electrochemical kinetics, and its kinetic constant is up to  $8.6 \times 10^{-2} \text{ cm s}^{-1}$  on the oxidative pretreated pyrographite electrode [27]. The kinetics of the Cr(II)/Cr(III) redox couple is slow on most the carbonaceous electrodes, while the kinetic constant of Cr(II)/Cr(III) redox couple on the electrodeposited Bi catalyst is as high as  $1.35 \times 10^{-3} \text{ cm s}^{-1}$  [28], which is generally comparable to other redox couples reported in the literature [29]. Moreover, the Bi catalyst has a high overpotential to suppress hydrogen evolution during the reducing process of Cr(III) to Cr(II). However, the Bi catalyst has a relatively low oxidation potential (0.05 V vs. SHE), and tends to be oxidized by air or ferric ions. To avoid this issue, the typical method for catalyst preparation is to add a small amount of  $\text{Bi}^{3+}$  ions in the electrolyte and synchronously electrodeposit the metallic particles on the electrode at the beginning of charge process [1,25,30]. During the in-situ electrodeposition process, the flow field design has great influence on the mass transport and concentration distribution of  $\text{Bi}^{3+}$  ions in the porous electrode, and further affects the catalyst distribution, which influences the active surface area and activation loss of the ICRFB. Previous studies have involved the effects of flow field designs on the mass transport characteristics in vanadium redox flow batteries (VRFBs) [22,31–34]. Shohji Tsushima *et al.* found that the VRFB with the interdigitated flow field (IFF) delivered a higher mass transport limitation than that with the SFF due to the enhanced convection of electrolyte in the electrodes [33]. In addition, other studies found that the IFF design has lower pump loss than the SFF design does [31,32].

Unlike the situations in VRFBs, the flow field designs in ICRFBs not only influence the transport and distribution of redox-active ions (Fe and Cr ions) in the porous electrode, but also affect the activation loss, as the Bi catalyst distribution is susceptible to the transport and concentration distribution of  $\text{Bi}^{3+}$  ions (mM level) during the in-situ catalyst electrodeposition process. In this work, the effects of flow field designs on the catalyst electrodeposition process and battery performance are investigated. It is further found that the energy efficiency of the ICRFB with the IFF reaches 80.7% at a high operating current density of  $320 \text{ mA cm}^{-2}$ , which is

8.2% higher than that of the ICRFB with the SFF.

## 2. Experimental

### 2.1. ICRFB setup

The lab-scale ICRFB with the IFF was designed and fabricated. The carbon papers (SGL, 10AA, 0.4 mm thickness) were pretreated in air at  $500 \text{ }^\circ\text{C}$  for 5 h and served as the electrodes. Both the negative and positive electrodes were made of two layers of carbon papers with active area of  $2.0 \text{ cm} \times 2.0 \text{ cm}$ , which were separated by the Nafion<sup>®</sup> NR-212 membrane. The polytetrafluoroethylene gaskets with 0.5 mm thickness were used to give an electrode compression ratio of approximately 40%. The IFFs with the channel depth 1.5 mm, the channel width 1.0 mm and rib width 1.0 mm were machined on the graphite plates. The gold-coated copper current collectors were adjacent to the graphite plates, and were clamped by the aluminum end plates. 20 mL mixed-reactant solutions of 1.0 M  $\text{FeCl}_2$  (Aladdin) + 1.0 M  $\text{CrCl}_3$  (Aladdin) + 3.0 M HCl (VWR) + 0.005 M  $\text{Bi}^{3+}$  ( $\text{Bi}_2\text{O}_3$ : Aladdin) were used as both the anolyte and catholyte. The electrolytes were circulated in the Norprene<sup>®</sup> #16 Chemical Tubing by a 2-channel peristaltic pump (Longer pump, BT100-1F). The SFF design, one of the most widely used flow field design in fuel cells and flow batteries [9,31,35–39], was used for comparison. For the ICRFB with the SFF, the components such as the channel, rib, electrode, gasket and membrane were identical with the ICRFB with the IFF.

### 2.2. Test and characterization

The cell tests were conducted on Arbin BT2000 (Arbin<sup>®</sup> Instrument). The cell and the electrolyte reservoirs were placed in a temperature chamber at  $65 \text{ }^\circ\text{C}$ . For the polarization test, the ICRFBs were charged to approximately 50% state-of-charge (SOC) before the tests. For each current density, the battery discharged for 20 s, rested for 10 s, and charged at the same current density for 20 s to keep the SOC constant during the polarization test. For the charge-discharge tests, the charge cut-off voltage was 1.2 V to mitigate hydrogen evolution, which could occur at the negative electrode during the charge process, and the discharge cut-off voltage was 0.8 V. The flow rate of the electrolyte was  $50 \text{ mL min}^{-1}$ . The cycle test was conducted at  $320 \text{ mA cm}^{-2}$  and  $65 \text{ }^\circ\text{C}$ , and 50 mL mixed-reactant solutions of 1.0 M  $\text{FeCl}_2$  + 1.0 M  $\text{CrCl}_3$  + 3.0 M HCl + 0.002 M  $\text{Bi}^{3+}$  were used as both the anolyte and catholyte.

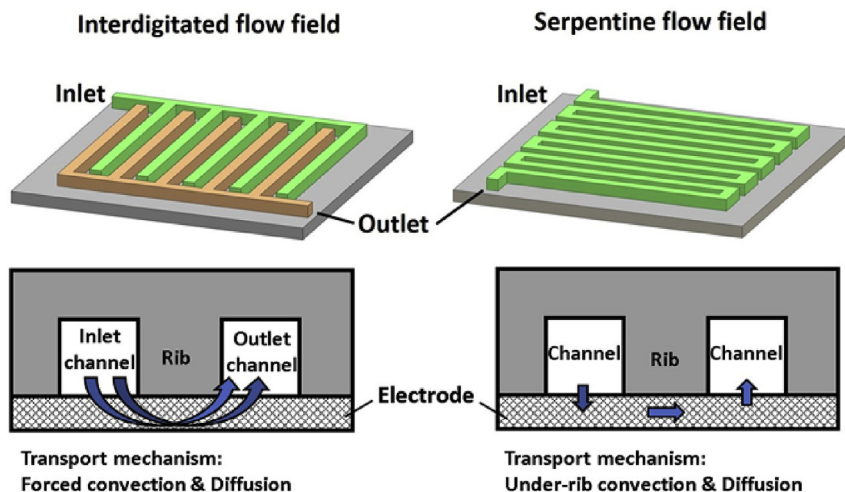


Fig. 1. Schematic of the interdigitated flow field and serpentine flow field.

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