



## Development of coin-type cell and engineering of its compartments for rechargeable seawater batteries



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### HIGHLIGHTS

- The cell design and component optimization for seawater batteries were performed.
- Engineering the cathode compartment improved the cell performance.
- Optimized coin-type cell can be used to test various types of electrochemical cell.

### ARTICLE INFO

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Cathode current collector  
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### ABSTRACT

Cell design and optimization of the components, including active materials and passive components, play an important role in constructing robust, high-performance rechargeable batteries. Seawater batteries, which utilize earth-abundant and natural seawater as the active material in an open-structured cathode, require a new platform for building and testing the cells other than typical Li-ion coin-type or pouch-type cells. Herein, we present new findings based on our optimized cell. Engineering the cathode components—improving the wettability of cathode current collector and seawater catholyte flow—improves the battery performance (voltage efficiency). Optimizing the cell component and design is the key to identifying the electrochemical processes and reactions of active materials. Hence, the outcome of this research can provide a systematic study of potentially active materials used in seawater batteries and their effectiveness on the electrochemical performance.

### 1. Introduction

Excessive emissions of CO<sub>2</sub> from the combustion of fossil fuels are the main causes of climate change and environmental pollution, which are the global issues to be solved for a sustainable society. These issues require the development of new types of systems and technologies for energy conversion and storage based on renewable energy [1,2]. The increasing demand for the efficient utilization of renewable energy has led to the development of various types of energy storage systems (ESSs) [3,4]. The main purpose of ESSs is to store renewable energy and use it with high energy efficiency on demand. Currently, lithium-ion batteries (LIBs) technology is a mature battery technology that holds a major position in the ESS market because of their high energy density (~300 Wh kg<sup>-1</sup>), long cyclability (≥2000 times), and competitive energy cost (250–400 \$ kWh<sup>-1</sup>) [4–6]. However, their long-term

sustainable use may be limited because of the steeply rising prices of Li-containing raw materials due to the increasing demand for LIBs in electric vehicles and large-scale ESSs and due to their finite geographical distribution [7].

Our group has recently introduced a novel, low-cost, and eco-friendly rechargeable seawater battery using earth-abundant, natural seawater as the active material [8–15]. Fig. 1a illustrates the schematic of the basic structure and the components of a seawater battery cell. The cell consists of two compartments, an anode and a cathode, which are separated by a NASICON ceramic electrolyte (Na<sub>3</sub>Zr<sub>2</sub>Si<sub>2</sub>PO<sub>12</sub>). The anode compartment is composed of a sodium metal anode attached to a current collector and a non-aqueous liquid electrolyte. The cathode part consists of a cathode current collector and seawater catholyte. Utilizing seawater that contains Na<sup>+</sup> and Cl<sup>-</sup> ions, the batteries operate based on the redox reactions of Na<sup>+</sup> ions at the anode side and the simultaneous

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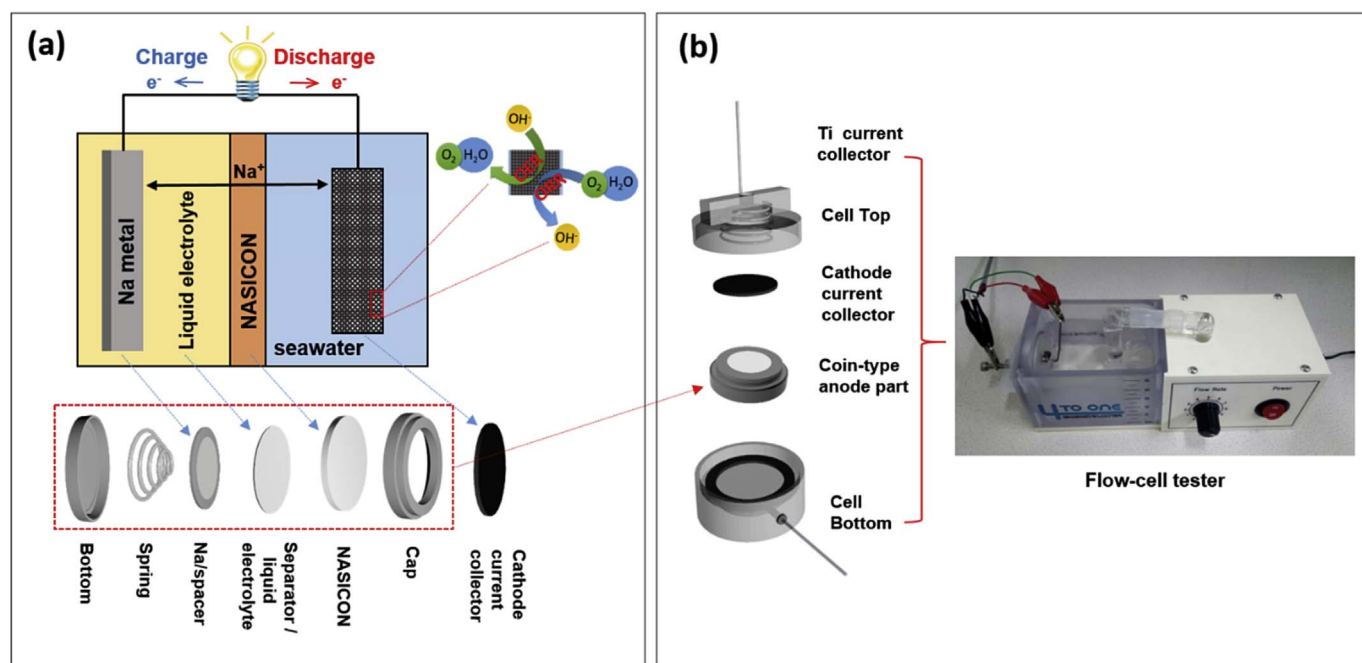
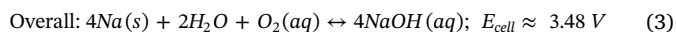
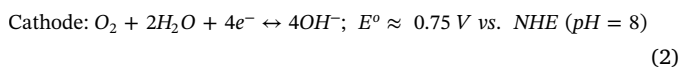
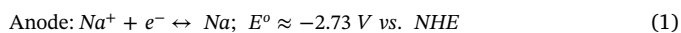


Fig. 1. (a) Schematic diagram of the seawater battery (top) and disintegrated cell components of the coin-type cell (bottom) and (b) assembled coin-type cell and flow-cell tester.

evolution/reduction reactions of gaseous  $O_2$  and  $Cl_2$  in seawater at the cathode side during charge and discharge processes. In principle, the oxygen evolution/reduction reaction (OER/ORR) is thermodynamically preferred over the chlorine evolution/reduction reaction (CIER/CIORR), according to the Pourbaix diagram of a water electrolyte containing  $Cl^-$  ions; however, the proportion of the two reactions depends on the operating conditions near the cathode current collector. Considering the pH of seawater ( $\sim 8$ ),  $Na^+$  ion content in seawater ( $\sim 0.47$  M) and oxygen partial pressure at 100% saturation from ambient air ( $\sim 0.2$  atm), the half-cell and full-cell reactions during the charge/discharge processes and the theoretical cell voltage ( $E_{cell}$ ) can be described as follows [11]:



During discharge, the Na metal anode is oxidized to  $Na^+$  ions and transported into the seawater catholyte through the NASICON membrane. At the same time, the ORR occurs, forming water-soluble NaOH at the cathode side. The cell is charged by the reduction of  $Na^+$  ions from seawater onto the anode in the opposite manner, while seawater oxidation (OER) occurs at the cathode side.

The seawater battery system employs multilayer electrolytes consisting of non-aqueous (anolyte) and aqueous (seawater catholyte) electrolytes and a ceramic electrolyte (NASICON) between them. Such a structural feature of the cell requires a new type of cell platform and a testing environment other than a typical 2032 coin-type cell. Previously, we have reported a comparative study between sodium, beta-alumina ( $Na, \beta''-Al_2O_3$ ), and  $Na_3Zr_2Si_2PO_{12}$  as the  $Na^+$  ion conducting solid electrolyte for seawater batteries [16]. We also investigated non-aqueous electrolytes at the anode side such as ether-based or ionic liquid electrolytes [10,17], as well as negative electrodes to replace the Na metal, and hence to achieve Na metal-free seawater batteries [8,10,14]. It was found that the cell design, the choice of the component material, and its engineering could affect the cell performance of the materials being investigated as potential electrodes and electrolytes. Hence, it is essential to have the normalized cell and its

standard testing condition so that the potential chemicals can be easily tested and their results can be compared to those obtained in the other labs. This will greatly contribute to the further development of seawater batteries by providing many choices of key materials. For example, the great success of the Li-ion battery technology also started with developing a coin-type cell design that allowed researchers to investigate and discover its key electrode and electrolyte materials.

In this work, we show the importance of cell components in seawater batteries and the optimized cell performance by engineering them and highlighting the significance of the cell design and component engineering. The effect of wettability of the cathode current collector was investigated. As a low-cost, highly conductive cathode current collector, a commercially available carbon felt was selected and the surface wettability was examined to improve the charge-discharge behaviors. In addition, the flow effect of the seawater catholyte was studied by conducting a comparison test of flow ON/OFF states. Furthermore, we proved the sluggish kinetics of the OER/ORR on the cathode current collector in seawater, which induces major kinetic limitations during the charge and discharge processes of seawater batteries, by comparing a seawater cell with a fast Na-ion-intercalating electrode material. To improve the cathode reaction kinetics, we employed several electrocatalysts facilitating the OER/ORR, highlighting significance of the role of the cathode current collector in achieving low-cost, high-performance seawater batteries.

## 2. Experimental methods

### 2.1. Preparation of solid electrolyte

NASICON ( $Na_3Zr_2Si_2PO_{12}$ ) was used as a solid electrolyte to separate the seawater cathode from the anode. The NASICON was fabricated by a solid-state reaction based on our previous study [9,10,18]. Briefly, the precursor powders of  $Na_3PO_4 \cdot 12H_2O$ ,  $SiO_2$ , and  $ZrO_2$  (Aldrich) were uniformly mixed using a mechanical ball mill and calcined at  $400$  °C and  $1100$  °C in ambient air. The calcined powder was ground and uniaxially pressed into pellets at  $7$  MPa. The pellets were sintered at  $1230$  °C for  $10$  h in ambient air. The diameter and thickness of final NASICON membranes were  $16$  mm and  $1.0$  mm, respectively, and the measured density was around  $3.07 \text{ g cm}^{-3}$  ( $\sim 94\%$  of the theoretical

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