

## Advanced rechargeable zinc-air battery with parameter optimization

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### HIGHLIGHTS

- Rechargeable zinc-air battery with a compacted structure was optimally designed.
- Electrochemical and structural performance of air electrode was characterized.
- Cycling performance of the battery was improved by electrolyte management.
- Oxygen bubbles movement was controlled by electromagnetic coupling.

### ARTICLE INFO

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### ABSTRACT

Zinc-air batteries will be a promising candidate for storage energy and power supply due to their high specific energy, environmental compatibility, and economic availability. However, the problem of cycle life of rechargeable zinc-air battery remains unresolved mainly because of dendrite growth of electrodeposited zinc and performance degradation of air electrode. Here we show that rechargeable zinc-air battery with an optimized structure can stably run at large current densities, where air electrode is connected to the charging electrode through a stainless steel frame, and the effective area of charging electrode is larger than that of zinc electrode by way of a trapezoidal structure. This battery structure can control morphological change of zinc electrode and monitor dendrite growth without increasing the battery volume. The results demonstrate that the charge-discharge efficiency of rechargeable zinc-air battery can be improved by nickel foam as gas diffusion layer of air electrode, calcium oxide additive to the electrolyte, or a permanent magnet in parallel with the electrode. The lifetime of rechargeable zinc-air battery can be extended by electrolyte flow or battery structure optimization. These findings will be available for other metal-air batteries and electrolytic metal industry.

### 1. Introduction

Zinc-air batteries have attracted more attention as one of energy storage devices in application of electric grid [1], green energy [2] and power supply [3] due to high-energy-density and non-pollution advantage. Nevertheless, the cycling performance of rechargeable zinc-air battery is unsatisfactory [4], where morphological change of electrodeposited zinc would shorten cycle life of the battery [5], and sluggish kinetics of oxygen redox reaction could decrease energy efficiency of the battery [6].

Energy efficiency of rechargeable zinc-air battery is mostly related to catalytic activation. Study of oxygen redox catalysts is mainly focused on precious metals and their alloys [7], carbon nanostructure materials [8], transition metal oxides [9], and inorganic/organic compound materials [10]. Moreover, the combination of transition metal oxide and carbon nanostructure can be available for improving the

comprehensive performance of oxygen redox reaction [11]. Unfortunately, rechargeable zinc-air battery with bifunctional catalysts [12,13] was generally applied at small currents in order to guard against material decomposition and catalyst loss. To meet fast-charge demand, a tri-electrode configuration was developed for rechargeable zinc-air battery [14], namely the charging and discharging proceed was separately employed with single electrode, and zinc electrode was located between air electrode and the charging electrode. Although this structure can avoid impairment of oxygen bubbles on the catalytic layer of air electrode, it would increase the battery volume. In addition, morphological change of zinc electrode would be more severe at large current densities, which can lead to short circuit of rechargeable zinc-air battery. To inhibit dendrite growth of electrodeposited zinc, many studies have been made in terms of electrolyte additives [15], electrode surfactant [16], and metal alloy [17], but these techniques would contaminate and even reduce active material of zinc electrode. Other

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### Nomenclature

Symbol Unit and value

$C$	concentration of zinc ion in the electrolyte, $\text{mol L}^{-1}$
$D$	diffusivity of zinc ion in the electrolyte, $\text{m}^2 \text{s}^{-1}$
$E$	potential, V
$f$	driving force, N
$F$	Faraday constant, $96,485 \text{ C mol}^{-1}$
$I$	local current density, $\text{A m}^{-2}$
$j$	charging/discharging current density, $\text{mA cm}^{-2}$
$L$	zinc electrode length, m
$p$	pressure, Pa

$P$	power device
$R$	resistance device
$S$	zinc ion reaction rate, $\text{mol L}^{-1} \text{s}^{-1}$
$v$	flow velocity, $\text{mL s}^{-1}$
$z$	valence of zinc
$Z_i$	imaginary part of electronic impedance, +2
$Z_r$	real part of electronic impedance
$\lambda$	mobility of the charged species
$\phi$	electrolyte potential, V
$\rho$	electrolyte density, $\text{kg m}^{-3}$
$\mu$	dynamic viscosity, $\text{N s m}^{-2}$

researchers have done much works in battery management in order to control morphological change of zinc electrode. Gavrilović-Wohl-muther et al. [18] analyzed morphology pattern of the zinc deposits at different electrolyte temperatures and various electrolyte flow velocities, demonstrating that electrolyte flow was an effective measure to inhibit dendritic morphology. Hwang et al. [19] developed a rechargeable zinc-air battery using commercial polypropylene membrane coated with polymerized ionic liquid as a separator, which can allow anionic transfer through the separator and minimize the migration of zincate ions to the cathode surface. Pichler et al. [20] employed pulse charging as suppressing dendrite growth of electrodeposited zinc, improving the cycling performance of rechargeable zinc-air battery. Increase of ion diffusion time is beneficial to control morphological change of electrodeposited zinc [21], whereas inefficient electro-deposition, increase of internal resistance and electrolyte pressure fluctuation would be accompanied by the above measures.

Additionally, performance degradation of air electrode becomes more prominent at high current densities, restricting cycle life of rechargeable zinc-air battery. Oxygen has paramagnetic property due to two unpaired electrons in the oxygen molecule, and thus the magnetic field can be applied to promote oxygen transfer. Wang et al. [22] stated that the catalytic activity of oxygen reduction reaction was enhanced under the condition of internal-external magnetic fields. Shi et al. [23] present that the magnetic field was used for improving discharging performance of zinc-air fuel cells. However, the effect of magnetic field on the charging performance of rechargeable zinc-air battery was rarely studied. What's more, oxygen bubble is easily adhered to the electrode surface, influencing stability of charging proceed.

In this work, rechargeable zinc-air battery with a compacted structure is proposed on the basis of bifunctional catalyst and tri-electrode configuration, where the charging electrode is touched with air electrode through the stainless steel framework, so the battery can be

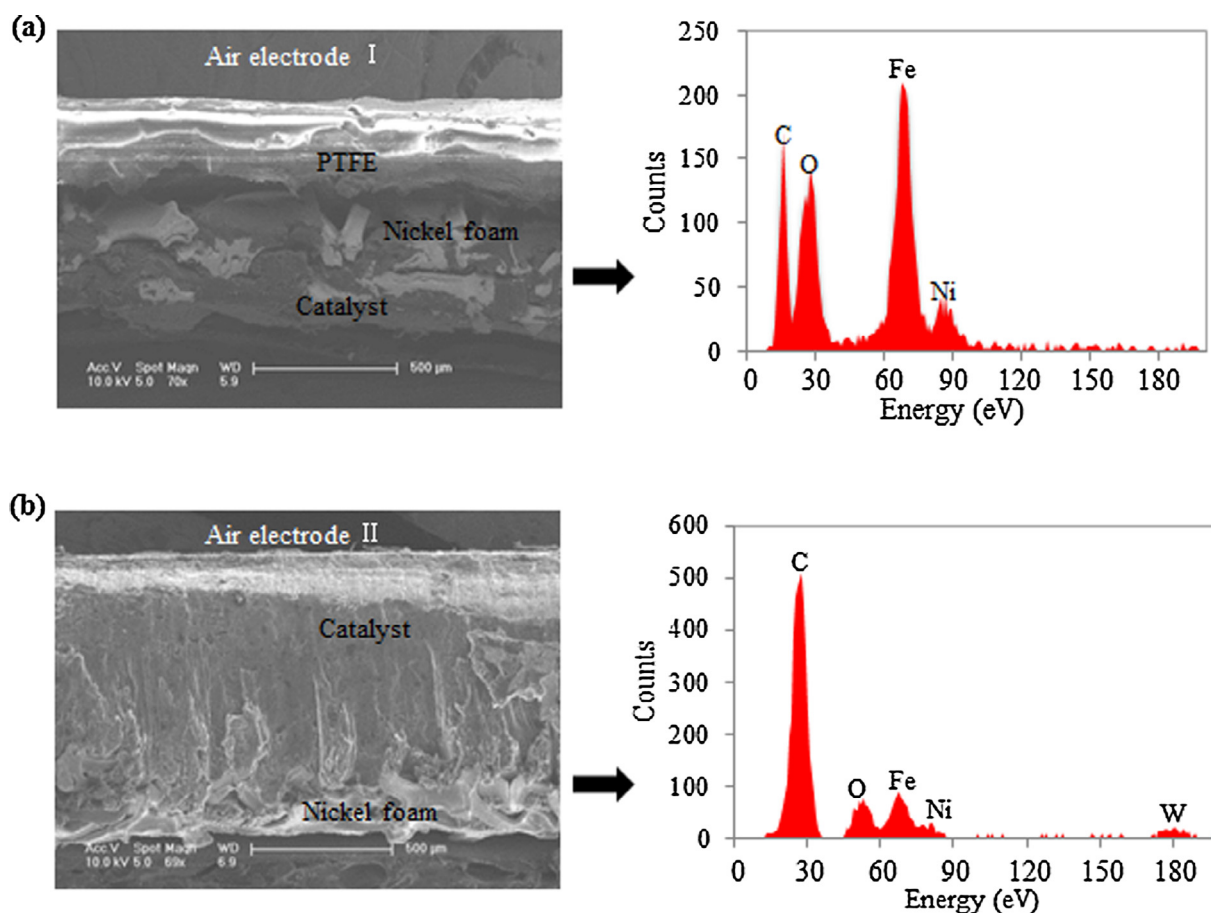


Fig. 1. Structure characteristics, (a) SEM image and X-ray spectrum of air electrode I, (b) SEM image and X-ray spectrum of air electrode II.

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