



Rechargeable urea-assisted Zn-air batteries: Concurrent fast-charging kinetics and contaminant urea removal

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Rechargeable zinc-air batteries (R-ZABs) have garnered significant attention as promising energy storage devices, attributed to their low cost, inherent safety, and good sustainability^[1]. Despite these advantages, the practical application of R-ZABs is hindered by the sluggish kinetics displayed by the oxygen reduction and evolution reactions (i.e., ORR and OER)^[2]. So far, efforts have focused on improving the discharge performance of R-ZABs through the use of high-activity ORR catalysts, such as metal-nitrogen-carbon (M-N-C), single/dual-atom catalysts (SACs/DACs), and high-entropy alloys^[3-5]. Unfortunately, the sluggish OER (1.23 V vs. RHE) during charging has been largely overlooked. Its high thermodynamic equilibrium potential not only escalates the energy input required for charging (often exceeding 1.9 V) but also leads to catalyst degradation, thereby shortening the useful lifespan of R-ZABs^[6].

To address this challenge, the OER in traditional R-ZABs can be replaced with favorable oxidation reactions of small molecules, such as urea, methanol, and ethanol^[7-9]. Among these alternatives, the urea oxidation reaction (UOR, $\text{CO}(\text{NH}_2)_2 + 6\text{OH}^- \rightarrow \text{N}_2 + \text{CO}_2 + 5\text{H}_2\text{O} + 6\text{e}^-$; 0.37 V vs. RHE) stands out as an ideal anode half-reaction for clean energy conversion and storage devices (Figure 1a,b)^[10-12]. The unique advantages of the UOR over other small-molecule oxidations are as follows: (i) compared to unstable and highly toxic hydrazine, urea is more stable in alkaline solutions and less toxic; (ii) urea is a common pollutant in wastewater. Utilizing the UOR not only reduces the charging potential but also purifies wastewater, a benefit that alcohol oxidation cannot provide. Nevertheless, the UOR suffers from torpid kinetics, involving multiple electron transfers, complex intermediate conversions, and CO poisoning^[13]. Therefore, it is highly crucial to develop high-activity and stable UOR catalysts.

Current research on UOR catalysts primarily focuses on three material categories: (i) Bulk Ni/Co-based catalysts, which attract extensive attention for their readily tunable 3d orbital electron configurations and high intrinsic activity^[14-16]. A self-supported Ag-CoOOH@NiOOH heterojunction, constructed by coupling an Ag-CoOOH nanosheet with a compact NiOOH layer, achieves high-energy interfacial structures^[16]. Such strong interactions enhance catalytic activity and selectivity for UOR, ensuring good stability. (ii) SACs, which maximize atom utilization efficiency and provide unique electronic properties for enhanced catalytic performance. For instance, Ni SAsNC exhibit near 100% atom efficiency and dual ORR/UOR functionality for urea-assisted R-ZABs^[12]. (iii) DACs, which optimize the adsorption and activation of urea molecules, thereby improving catalytic activity and selectivity. For example, Co-Ni DACs synergistically combine the properties of two different metals to show promise in the UOR process^[17]. Despite this progress, current research is still in the development stage regarding the regulation of the microscopic geometric structure of catalyst active sites for efficient UOR. Compared to traditional methods (e.g., doping, alloying, and constructing heterojunctions), curvature-induced intrinsic microstrain enables more precise and tunable regulation of metal center electronic structures, offering fine-tuning of the coordination environment around the active metal centers and effective modulation of their electronic properties^[18,19].

In a recent study, Kang and co-workers developed Fe SAs anchored on a highly curved NC dodecahedron with a concave morphology (Fe SA/NhC)^[20], which served as a superior bifunctional catalyst for both the ORR and UOR in urea-assisted R-ZABs. Curvature engineering has been proven to enable precise geometric and electronic control, directly manipulating bond lengths and coordination geometries of metal centers, which is crucial for optimizing catalytic activity. Additionally, the three-dimensional



curvature enhances mass transport and increases accessible active site densities, significantly boosting catalytic efficiency.

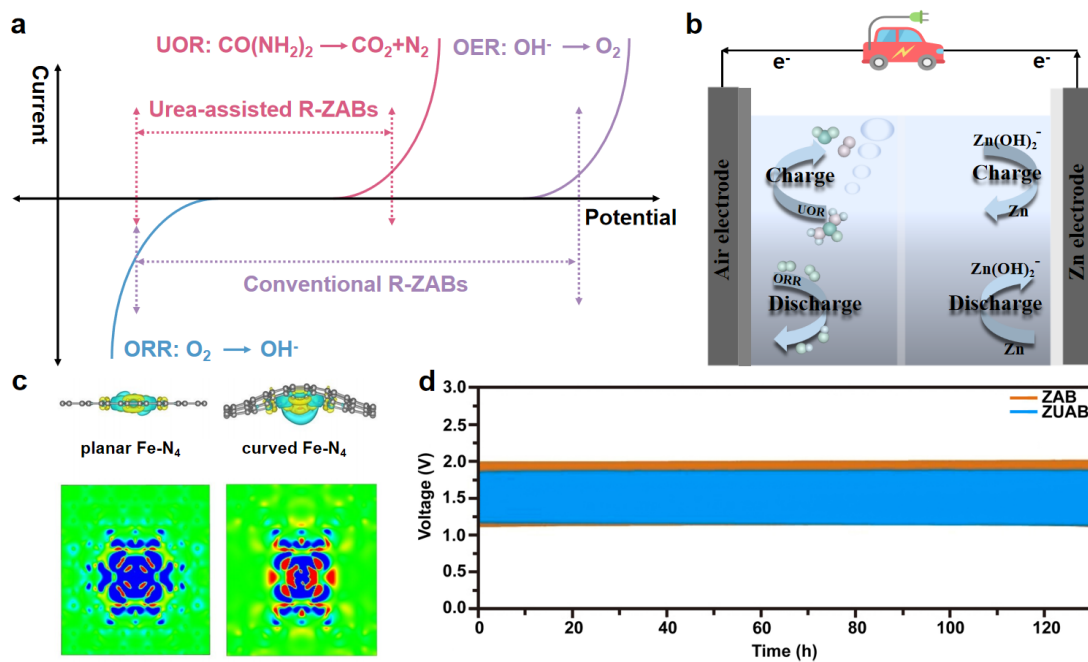


Figure 1. (a) Comparison of the potential gap between the anodic and cathodic reactions in conventional and urea-assisted R-ZABs; (b) Schematic diagram of urea-assisted R-ZABs. R-ZABs: Rechargeable zinc-air batteries.

By comparing SEM and TEM images, the morphology of Fe SA/NC differs significantly from that of Fe SA/NhC, revealing the presence of curved surfaces (Figure 2a,b,c,d). This is supported by extended X-ray absorption fine structure (EXAFS) data. The average Fe-N bond length in Fe SA/NhC was slightly shorter than that of Fe SA/NC (i.e., 1.92 vs. 2.01 Å, Figure 2e), indicating the formation of a local strain effect induced by the curved carbon nanostructure. Such high surface curvature of the carbon support facilitates fine-tuning of the spatial charge redistribution at Fe-N₄ sites (Figure 2f). Moreover, the curved Fe-N₄ site with moderate curvature was proven to show superior intrinsic ORR/UOR activity relative to the zero-strain planar Fe-N₄ site. The resultant urea-assisted R-ZAB prepared with Fe SA/NhC exhibited an ultra-low charge-discharge voltage difference of 0.686 V, significantly lower than that of conventional R-ZABs. It also exhibits long-term cycling stability for 130 h at 20 mA cm⁻².

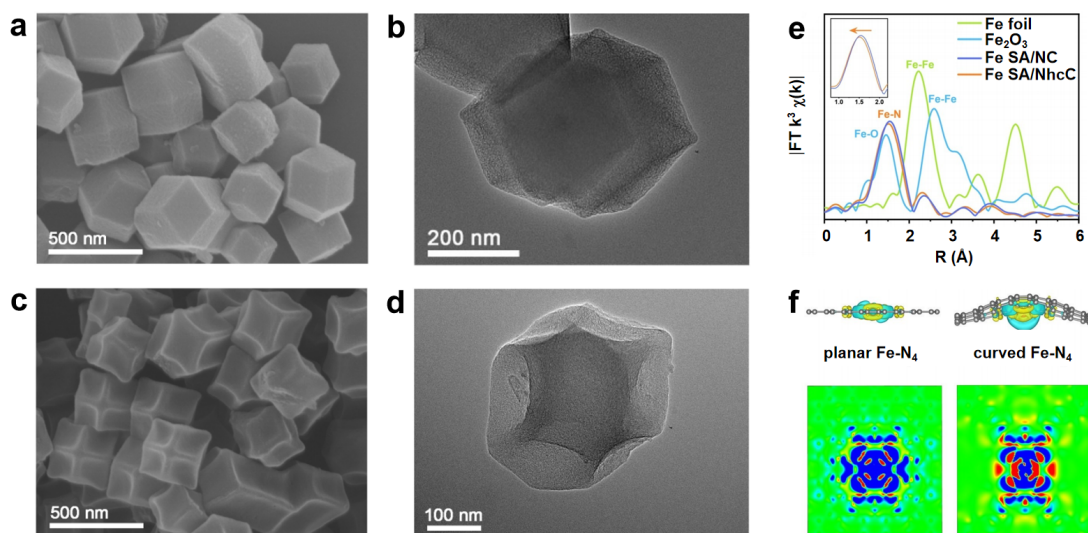


Figure 2. (a-d) SEM and TEM images of (a, b) Fe SA/NC and (c, d) Fe SA/NhC; (e) FT spectra as obtained from the k^2 -weighted Fe K-edge EXAFS data recorded for Fe SA/NC, Fe SA/NhC, Fe foil and Fe₂O₃; (f) Charge density difference (up) and 2D contour plot (down) for planar and curved Fe-N₄. Reproduced with permission from reference^[20]. Copyright © 2025 Wiley. SEM: scanning electron microscopy; TEM: transmission electron microscopy; SA: single atom; NhC: highly curved N-doped carbon dodecahedron with concave morphology; FT: fourier Transform; EXAFS: extended X-ray Absorption fine structure.

In summary, this work provides valuable insights into enhancing the catalytic activity of SACs through modulating the surface curvature and the geometric configuration. Importantly, the as-constructed urea-assisted R-ZABs catalyzed by Fe SA/NhC offer a new approach for concurrent fast-charging kinetics and contaminant urea removal. Looking ahead, several critical aspects warrant further exploration. (i) The scalability of SAC synthesis is essential for practical applications. Current synthesis methods often face challenges in maintaining the high dispersion and stability of SAs when scaled up. To address this, potential strategies such as template-free synthesis or continuous flow reactors can be employed to achieve scaled-up production of SACs while maintaining single-atom dispersion. (ii) Actual industrial wastewater contains a myriad of impurities that may interfere with the UOR process and affect catalyst stability. Future research should focus on enhancing the robustness of catalysts to withstand such complex environments, potentially through surface modification (e.g., functionalization with hydrophobic groups, grafting of polymeric layers) or the development of protective coatings (e.g., carbon encapsulation, atomic layer deposition). (iii) The integration of these catalysts into large-scale battery devices poses significant engineering challenges. Efficient electrode design, optimized mass transport, and system integration are crucial for realizing the full potential of urea-assisted R-ZABs in real-world scenarios. Key engineering priorities include the development of scalable electrode architectures and the optimization of electrolyte flow to ensure efficient mass transport.

Declarations

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AI-assisted tools were used for language editing only. The authors are responsible for the accuracy and scientific content of the article.

Authors contribution

Gong J: Data analysis, investigation, validation, writing-original draft, writing-review & editing.

Meng T: Data analysis, investigation.

Lv XW, Gong J: Conceptualization, project administration, supervision, validation, writing-review & editing.

All authors have given approval to the final version of the manuscript.

Conflicts of interest

Jianxin Gong is an Editorial Board Member of *Smart Materials and Devices*. The other authors declare no conflicts of interest.

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Consent for publication

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