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Electrolyte-in-Gas Architecture: A Platform for Atmospheric Open and Rechargeable Metal–Air Batteries

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Abstract

Metal–air batteries are promising energy-storage systems; however, their performance is often constrained by sluggish oxygen transport and inefficient gas–liquid–solid interfacial reactions in conventional flooded-cell architectures. Here, we introduce an atmospheric open architecture for rechargeable metal–air batteries based on an electrolyte-in-gas configuration, in which a continuous supply of aerosolised electrolyte microdroplets replaces the conventional bulk liquid electrolyte. Using Zn–air batteries as a representative system, we show that this architecture renews dynamic triple-phase boundaries, enhances oxygen transport, and enables reversible battery operation under open atmospheric conditions. Molecular simulations, computational fluid dynamics, and high-speed imaging collectively reveal the mechanisms governing aerosol transport and interfacial electrochemistry. Reversible oxygen evolution and ZnO redox reactions verify the feasibility and stability of rechargeable operation. Collectively, these results establish a distinct atmospheric open architecture for rechargeable metal–air batteries and provide a general design framework for atmospheric open gas-involved electrochemical energy-storage systems.

Gas-involved electrochemical systems are emerging as important platforms for sustainable energy conversion and storage. Technologies including fuel cells, water electrolysis, CO₂ conversion, and metal–air batteries depend on electrochemical processes in which gaseous species serve as reactants or products. Despite their broad potential, their performance is often constrained by inefficient gas transport and sluggish gas–liquid–solid electrochemical reactions.¹⁻⁶

Metal–air batteries are a representative class of gas-involved electrochemical systems that directly utilise atmospheric oxygen at air-breathing electrodes.⁷⁻¹⁰ Their high theoretical energy densities and favourable material sustainability make them promising candidates for next-generation energy storage.¹¹⁻¹⁵ Among them, aqueous Zn–air batteries (ZABs) are one of the most technologically mature systems, and provide a suitable model for investigating gas-involved electrochemistry because of their well-established reaction chemistry and reversible oxygen electrochemistry.¹⁶⁻¹⁹

The gas-conversion reactions occurring in air-breathing batteries are limited by both sluggish interfacial charge-transfer kinetics and inefficient transport of gaseous reactants through liquid electrolytes. The oxygen reduction and evolution reactions (ORR/OER) proceed through complex multielectron pathways involving O–O bond cleavage and formation, imposing intrinsically high activation barriers and substantial overpotentials. Accordingly, extensive efforts have focused on developing highly active electrocatalysts that accelerate oxygen conversion and improve interfacial charge transfer. Nevertheless, even with major advances in catalyst design, limitations associated with gas mass transport remain a persistent bottleneck to overall electrochemical performance.

ORR/OER are typical gas–liquid–solid triple-phase reactions that occur at triple-phase boundaries (TPBs), where gaseous oxygen, liquid electrolyte, and solid electrodes are

simultaneously present. Prior to charge transfer, oxygen must first reach the reaction interface through the electrolyte and adsorb onto the catalyst surface. Although liquid electrolytes are indispensable for ionic conduction and the establishment of electrochemical interfaces, the low solubility and diffusivity of oxygen in liquids severely impede gas transport.^{4,20,21} In conventional flooded electrodes, oxygen diffusion through the electrolyte can therefore become rate-limiting, suppressing the formation and continuous renewal of effective TPBs and ultimately compromising electrochemical performance.^{22,23} Strategies such as forced convection, active gas supply, and optimisation of electrode and gas-diffusion-layer structures have been explored to alleviate these limitations, yet an efficient, practical, and scalable solution remains lacking.^{12,24-29}

To address the inherent mass-transport limitations of conventional gas-involved electrochemical systems, we previously developed an atmospheric open electrochemical architecture based on an aerosolised electrolyte, termed the “aeroelectrolyte”.³⁰ In this electrolyte-in-gas configuration, aeroelectrolyte microdroplets are continuously generated from a bulk solution and delivered directly to the electrode surfaces, thereby replacing the conventional flooded-electrolyte environment (**Supplementary Fig. 1**).³¹⁻³³ The resulting architecture provides direct gas access to the electrochemical interface, promotes continuous renewal of TPBs, efficient release of gaseous products, and lean-electrolyte operation. Ionic conduction between the electrodes is simultaneously maintained through a porous nanofibrous salt bridge. Although this architecture was previously validated in a primary Daniell cell, its applicability to reversible electrochemistry and rechargeable metal–air batteries has not yet been established.

In this study, we extend the electrolyte-in-gas concept to rechargeable Zn–air batteries and establish an atmospheric open architecture for reversible ZAB operation (**Fig. 1**). Using ZABs as a model system, we systematically optimise key design parameters, including electrode

configuration, electrolyte composition, and aerosol-spraying conditions. High-speed imaging, molecular simulations, and computational fluid dynamics are further combined to clarify aerosol transport behavior and interfacial electrochemistry, including the distinct solvation characteristics and localised pH regulation associated with aeroelectrolyte microdroplets. We subsequently demonstrate reversible ZAB operation under open atmospheric conditions. Together, these findings provide a mechanistic basis and design framework for developing rechargeable air-breathing electrochemical energy-storage systems based on electrolyte-in-gas architectures.

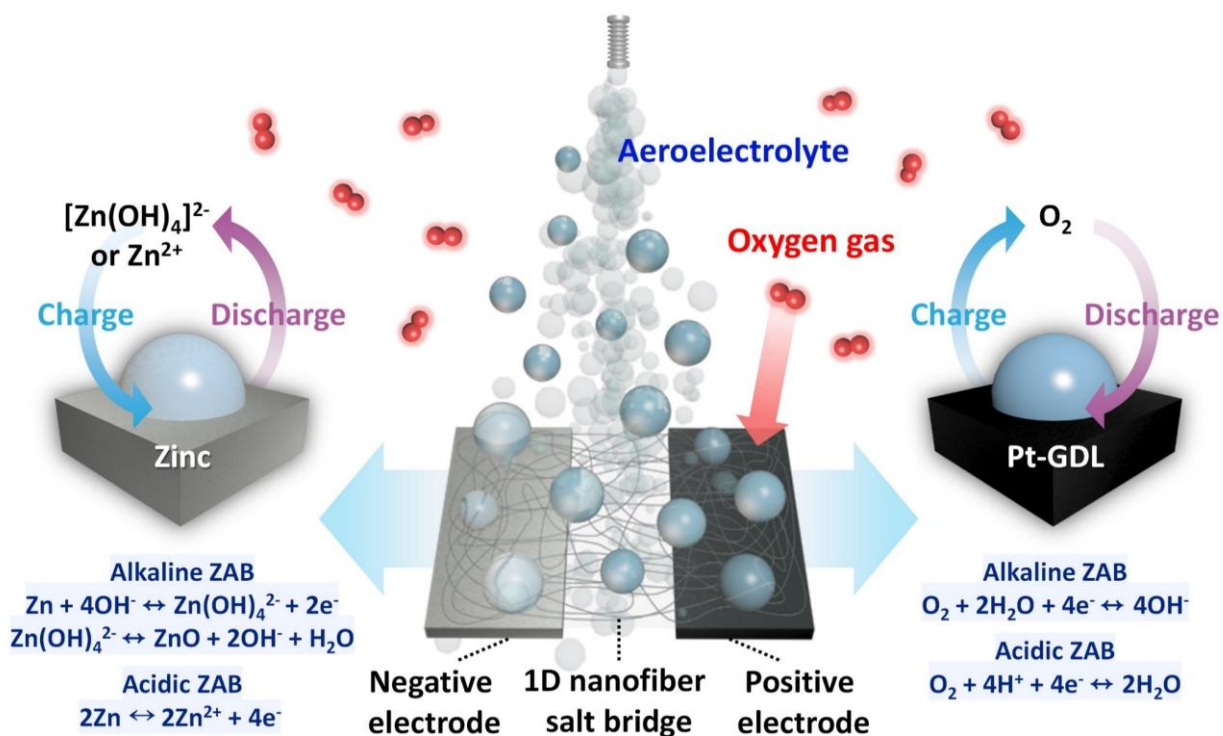


Fig. 1 | Schematic illustration of the aeroelectrolyte system for a Zn–air battery

RESULTS AND DISCUSSION

Customised cell design for realising atmospheric open metal–air batteries

A conventional aqueous Zn–air battery comprises a zinc negative electrode, an air positive electrode, and a bulk liquid electrolyte that provides ionic conduction between the two electrodes (**Fig. 2a**). The electrochemical reactions involve zinc stripping/plating coupled with ORR/OER, with distinct reaction pathways in alkaline and acidic electrolytes.^{34–37} In conventional Zn–air batteries, the electrodes are typically arranged in a face-to-face configuration and separated by the bulk liquid electrolyte. Although the electrolyte is essential for ion transport and electrochemical reactions, it also creates an inherent mass-transport barrier because of the low solubility and diffusivity of oxygen in aqueous media (**Fig. 2b and Extended Data Fig. 1**). Under standard conditions (25 °C, 1 atm), the solubility of oxygen in water is only ~1.2 mM, resulting in insufficient oxygen availability at the electrochemical interface.³⁸ During discharge, continuous oxygen consumption often requires forced gas transport through gas-diffusion layers or external gas bubbling to sustain the reaction. During charging, by contrast, evolved oxygen can accumulate as bubbles on the electrode surface, blocking active sites and disrupting continuous electrochemical operation. These mass-transport limitations become increasingly pronounced at high current densities, ultimately limiting reaction kinetics and overall battery performance.^{26,39,40}

To overcome these structural and mass-transport limitations, we developed an atmospheric open electrochemical cell architecture in which the electrolyte is continuously supplied to the electrode surfaces as fine aerosol droplets rather than filling the entire interelectrode space as a bulk liquid (**Fig. 2c**). This electrolyte-in-gas architecture spatially decouples gas transport from ion transport, allowing gaseous reactants and products to directly access or leave the electrochemical interface through the surrounding atmosphere while preserving localised ionic conduction. Consequently,

gas–liquid–solid TPBs are continuously formed and renewed, thereby alleviating the mass-transport limitations inherent to conventional bulk-electrolyte systems.

To accommodate the aerosolised electrolyte, the electrode configuration was redesigned from the conventional face-to-face arrangement to a parallel geometry aligned with the spray direction, thereby maximising interactions between aereoelectrolyte microdroplets and the electrode surfaces (**Fig. 2c**). Replacing the bulk electrolyte with an open atmosphere, however, disrupts continuous ionic transport between the two electrodes.^{41,42} To restore ionic conduction while maintaining open access to aerosol droplets and gaseous species, a porous nanofibrous salt bridge was introduced to connect the positive and negative electrodes. The salt bridge was fabricated by electrospinning polyacrylonitrile (PAN) and polyethylene oxide (PEO), providing mechanical robustness and ionic conductivity, respectively.^{43,44} This design enables localised electrochemical reactions within aereoelectrolyte microdroplets while maintaining continuous ionic transport between the electrodes, thereby supporting stable operation of the atmospheric open cell.

To optimise the salt bridge, cells were fabricated using electrospinning times of 0, 1, 10, and 30 min, and the influence of nanofiber density on electrochemical performance was systematically investigated. SEM images confirmed the progressive formation of porous nanofiber networks on the electrode surfaces with increasing electrospinning time, whereas no interconnected fibrous structure was observed in the non-electrospun cell (**Fig. 2d, Extended Data Fig. 2 and Supplementary Fig. 2**). The cell without a salt bridge exhibited a low and unstable open-circuit voltage (OCV), together with negligible current generation, reflecting the absence of a continuous ionic conduction pathway (**Fig. 2e,f**). By contrast, all electrospun cells displayed stable operating voltages and measurable current outputs, demonstrating that the nanofibrous salt bridge effectively restored ionic transport between the electrodes. Among these cells, the 10-min electrospun cell

delivered the highest voltage and current, representing an optimal balance between ionic conduction and electrolyte accessibility. Shorter electrospinning times provided insufficiently developed ionic pathways, whereas excessive fibre deposition at 30 min reduced network porosity and limited aeroelectrolyte penetration and electrode wetting. Consequently, the overcoated electrode exhibited gradual performance degradation during prolonged operation, likely because of restricted interfacial transport and the accumulation of reaction products. These results demonstrate that both the presence and porosity of the salt bridge are critical for sustaining efficient electrochemical operation in the electrolyte-in-gas architecture.

We next investigated how electrode–electrolyte interfacial characteristics affect electrochemical performance in the electrolyte-in-gas architecture. Because the aeroelectrolyte droplets exhibited a narrow size distribution under the present operating conditions, variations in cell performance could be attributed primarily to the properties of the electrode surfaces. Four electrode materials—gas diffusion layer (GDL), nickel foam, nickel mesh, and nickel foil—were evaluated under otherwise identical cell configurations. Contact-angle measurements showed progressively improved wettability in the order $\text{GDL} < \text{Ni foam} \approx \text{Ni mesh} < \text{Ni foil}$ (**Extended Data Fig. 3a**). This trend was consistent with the surface morphologies observed by SEM: the highly porous GDL retained air pockets that impeded droplet spreading, whereas the smooth Ni foil promoted more uniform electrolyte wetting and a larger effective electrode–electrolyte contact area.^{45–47} Consequently, electrochemical performance followed the same general trend. The GDL-based cell exhibited the largest overpotential, while the Ni foil electrode delivered the highest operating voltage and current; Ni foam and Ni mesh showed intermediate performance (**Extended Data Fig. 3b,c**). These results indicate that improved electrode wettability and interfacial contact are important for maximising electrochemical activity in the electrolyte-in-gas system.

To further reduce the activation overpotential, the Ni foil positive electrode was replaced with platinum-loaded carbon paper (Pt-GDL), which provides superior catalytic activity toward the ORR.^{48,49} The Pt-GDL electrode significantly enhanced oxygen-reduction kinetics, increasing the open-circuit voltage to ~1.2 V and the reaction current to ~18 μA (**Fig. 2h,i**). Together, these results demonstrate that optimising both electrode–electrolyte interfacial contact and ORR catalytic activity is essential for achieving high-performance atmospheric open ZABs.

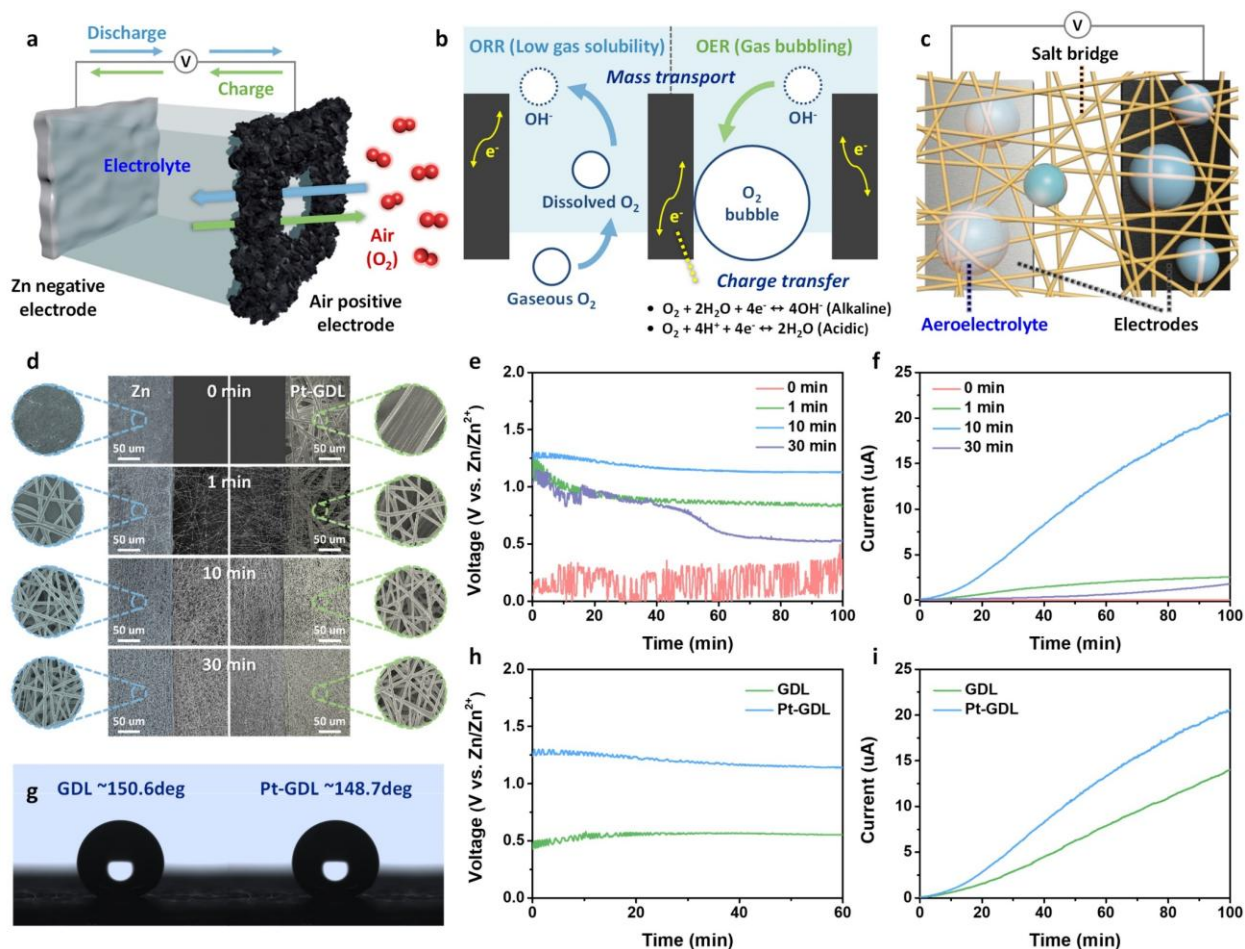


Fig. 2 | Aeroelectrolyte cell design. **a–c**, Schematics of (a) a conventional Zn–air battery, (b) its kinetic limitations, and (c) the aeroelectrolyte-based Zn–air battery configuration. **d**, SEM images of electrodes with salt bridges electrospun for 0, 1, 10, and 30 min (top to bottom). **e,f**, Electrochemical performance at different electrospinning times: (e) cell voltage profiles and (f) corresponding current profiles. **g**, Contact-angle measurements of the electrolyte on GDL and Pt-GDL electrodes. **h,i**, Electrochemical performance of GDL electrodes with and without Pt catalyst: (h) cell voltage profiles and (i) current profiles.

Local ion environment of aeroelectrolyte droplets

Although aerosolisation is generally regarded as a physical process that disperses a bulk electrolyte into fine droplets, it fundamentally reconstructs the chemical environment at the gas–liquid interface.⁴⁰ Surprisingly, bulk 6 M KOH (pH $\approx$ 14, black) exhibited a pronounced decrease in apparent surface pH to approximately 9 (brown) immediately after aerosolisation, even in the absence of electrochemical reactions, as confirmed by litmus paper measurements (**Fig. 3a**). This observation suggests that individual aeroelectrolyte droplets undergo substantial interfacial ion redistribution rather than simply preserving the local chemical environment of the bulk electrolyte.

To elucidate the origin of this behavior, molecular dynamics (MD) simulations were performed using a symmetric slab model consisting of a 6 M KOH aqueous phase confined between gas phases (N₂, O₂, or air), while an otherwise identical bulk-liquid system without a gas phase served as the control (**Extended Data Fig. 4**).^{40,50,51} In the bulk-liquid model, K⁺, OH[−], and H₂O remained relatively uniformly distributed throughout the solution. By contrast, introduction of a gas–liquid interface induced rapid molecular rearrangement H₂O preferentially accumulated near the interface, whereas OH[−] was depleted from the interfacial region within 20 ns (**Fig. 3c–e and Extended Data Figs. 4 and 5**).

This interfacial ion redistribution originates from the preferential orientation and accumulation of polar H₂O molecules at the gas–liquid interface, which energetically disfavour the presence of hydroxide ions in the outermost surface layer. Consequently, an OH[−]-depleted interfacial region develops, lowering the local pH at the droplet surface where electrochemical reactions occur despite the unchanged overall composition of the electrolyte.

Quantitative evidence for this interfacial restructuring was obtained by examining the width of the OH[−]-rich core within the simulated slab. Relative to the control system ($\sim$ 52 Å), the width of

the high-concentration OH^- region decreased to ~ 41 Å in the aerelectrolyte model containing gas–liquid interfaces, with the largest reduction observed under ambient-air conditions (**Fig. 3b**). This contraction reflects the preferential accumulation of H_2O and simultaneous depletion of OH^- at both interfaces, effectively narrowing the bulk-like interior of the electrolyte. Importantly, the same overall trend was reproduced for N_2 , O_2 , and air, indicating that the interfacial reconstructing primarily originates from the presence of the gas–liquid interface rather than from the specific chemical identities of the gas phase.

These molecular-level findings are consistent with the experimentally observed decrease in surface pH (**Fig. 3a**) and demonstrate that the aerelectrolyte constitutes an interfacially dominated reaction medium that differs fundamentally from the corresponding bulk electrolyte. This unique local interfacial environment provides a mechanistic basis for understanding pH-dependent Zn–air reaction pathways, the nonlinear concentration dependence of ZnCl_2 electrolytes, and the further decrease in local alkalinity during charging.

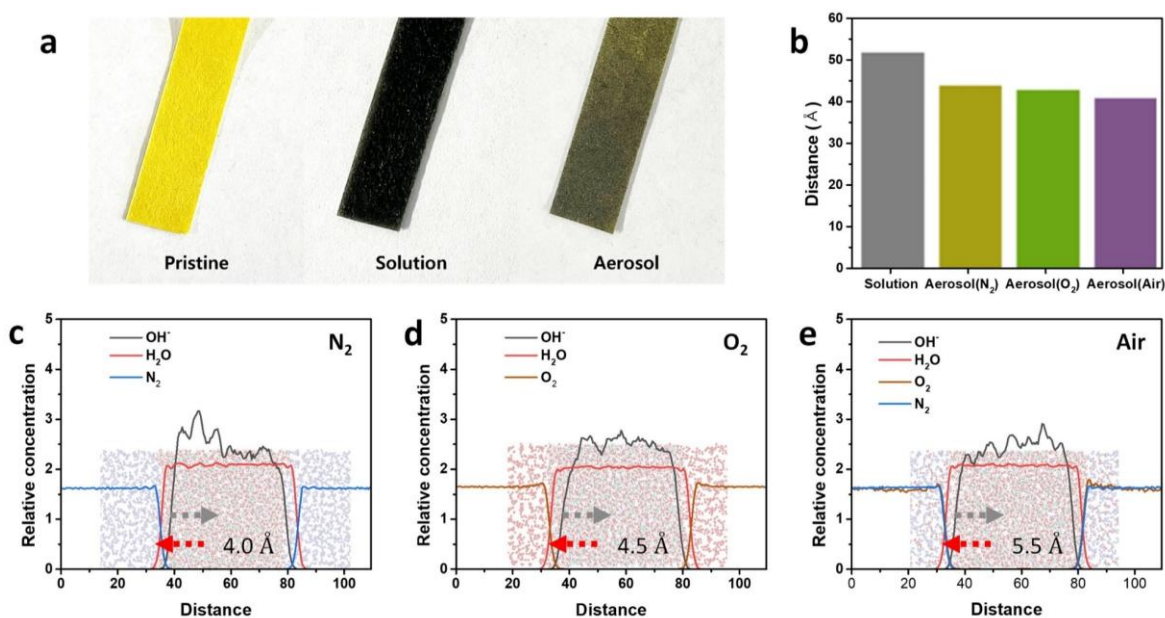


Fig. 3 | Interfacial pH distribution and gas-dependent molecular dynamics analysis of the aereoelectrolyte. a, pH indicator-paper results under pristine, solution, and aerosol conditions. **b**, Thickness of the OH⁻-rich region near the gas–liquid interface. **c–e**, Molecular dynamics simulation results showing relative concentration profiles at the gas–liquid interface under (c)N₂, (d) O₂, and (e) air environments.

Electrochemical optimisation and visualisation of aeroelectrolyte dynamics

To clarify the roles of oxygen supply and electrolyte concentration in the electrolyte-in-gas architecture, electrochemical measurements were combined with high-speed visualisation of aeroelectrolyte dynamics (**Fig. 4**). Electrolytes were first purged with N₂ or O₂ for 5 min before aerosolisation, followed by open-circuit voltage (OCV) and discharge current measurements. The aerated electrolyte produced higher and more stable OCV and current responses than the deaerated electrolyte (**Extended Data Fig. 6a–c**). Notably, despite its reduced dissolved oxygen content, the deaerated aeroelectrolyte still exhibited responses comparable to those of the conventional closed cell. High-speed imaging further showed similar aeroelectrolyte transport regardless of the dissolved gas species (**Extended Data Fig. 6d–g**). These observations suggest that dissolved oxygen contributes to cell performance but is not primary oxygen source ; instead, the open architecture enables continuous oxygen supply from the surrounding gas atmosphere, providing a distinct mass-transport advantage over conventional flooded-cell configurations.

Electrochemical performance was strongly affected by spray pressure and electrolyte concentration. At a fixed ZnCl₂ concentration, increasing the spray pressure from 0.1 to 0.25 MPa raised the operating voltage from ~0.8 to ~1.2 V and discharge current from ~3.7 to ~20.5 μ A (**Fig. 4a–c**). High-speed imaging revealed that higher pressure generated denser, faster aeroelectrolyte streams, enhancing droplet delivery and interfacial renewal at the electrode (**Fig. 4g, Extended Data Fig. 7 and Supplementary Video 1**). These pressure-induced flow dynamics enhanced mass transport and stabilised electrochemical operation. Electrolyte concentration also strongly influenced performance (**Fig. 4d–f and Extended Data Fig. 8**). Although higher ZnCl₂ concentrations promoted aeroelectrolyte generation and transport performance exhibited a non-monotonic dependence on concentration, peaking at 3 M rather than 5 M. This behavior reflects

the competition between improved aerelectrolyte transport and concentration-dependent electrolyte chemistry. Increasing concentration shifted the air-electrode potential positively as pH decreased, thereby increasing cell voltage (**Fig. 4f,i and Extended Data Fig. 9**). However, at 5 M, excessive droplet accumulation caused local flooding, hindered oxygen transport, and destabilised gas–liquid–solid TPBs. Near-saturation conditions may also restrict ion mobility and Zn dissolution. Consequently, 3 M ZnCl_2 provided the optimal balance between mass transport and electrochemical reactivity in the electrolyte-in-gas architecture.

To complement the high-speed imaging analysis, computational fluid dynamics (CFD) particle-tracking simulations were performed using the experimental reactor geometry and aerosol injection conditions. The simulations reproduced the experimentally observed flow behavior, increasing the spray pressure intensified internal convection and enhanced aerelectrolyte transport toward the electrode region (**Fig. 4h**), whereas, increasing ZnCl_2 concentration increased local droplet density and recirculation around the electrode (**Fig. 4j**). These results indicate that the superior performance at 3 M ZnCl_2 originates from efficient droplet transport and continuous electrolyte replenishment at the gas–liquid–solid TPBs. At 5 M, however, the higher simulated droplet density supports the experimentally inferred onset of excessive interfacial wetting rather than further enhancement of effective TPB formation. The CFD analysis therefore provides mechanistic support for the experimentally observed optimum electrolyte concentration in the atmospheric open metal-air battery architecture.

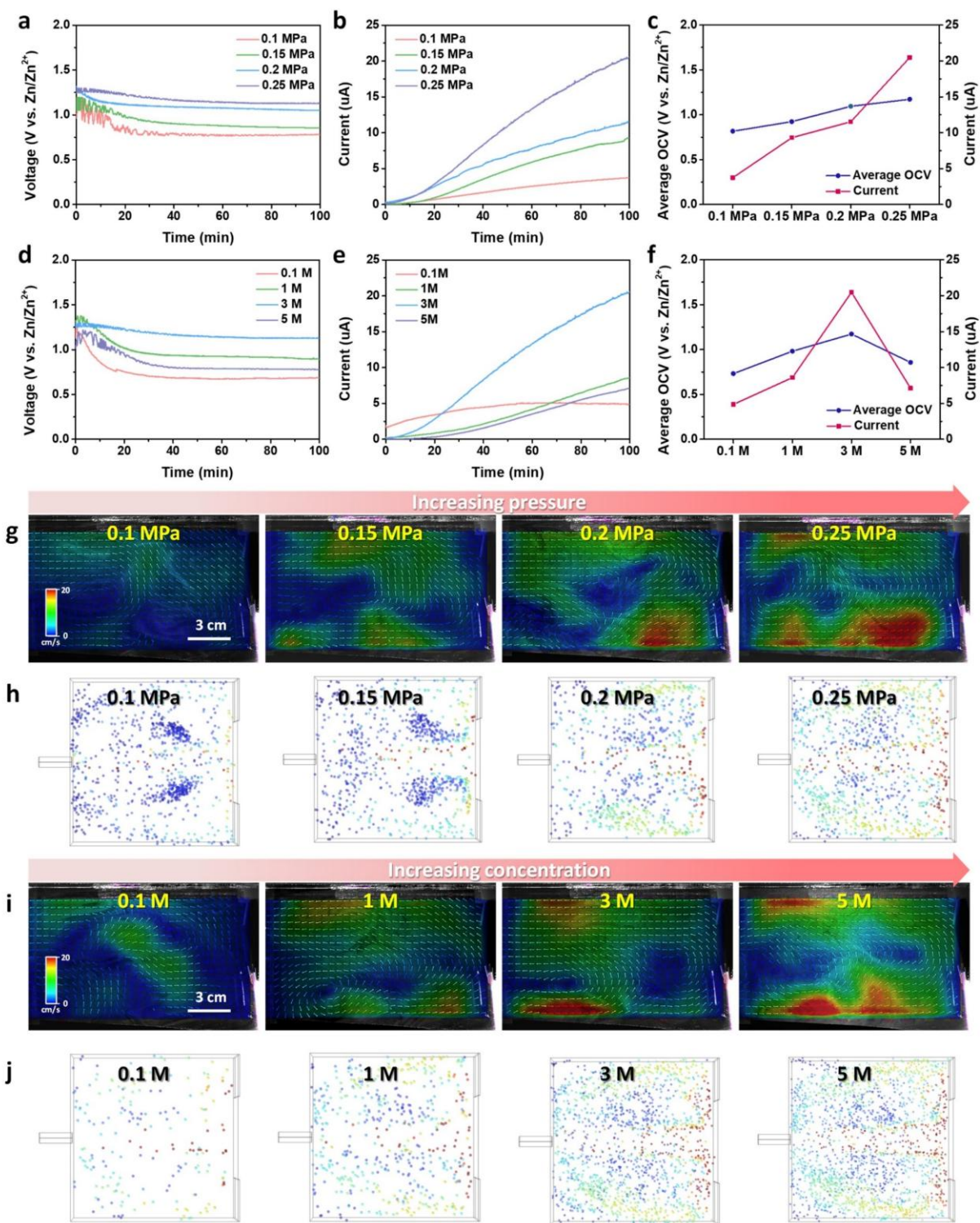


Fig. 4 | Optimisation of aeroelectrolyte parameters and high-speed imaging of flow dynamics and convection behavior. **a–c**, Electrochemical performance at constant O₂ purging and ZnCl₂ concentration (3 M) with varying aerosol-generation pressure: (a) cell voltage profiles, (b) current profiles, and (c) average OCV and current values. **d–f**, Electrochemical performance at constant O₂ purging and pressure (0.25 MPa) with varying ZnCl₂ concentration: (d) cell voltage profiles, (e) current profiles, and (f) average OCV and current values. **g,h**, Flow-rate analysis from (g) high-speed imaging and (h) dynamic simulations under different aerosol-generation pressures. **i,j**, Flow-rate analysis from (i) high-speed imaging and (j) dynamic simulations under different ZnCl₂ concentrations.

Atmospheric open Zn-air battery reaction mechanisms in different electrolytes

As ZAB reaction pathways are strongly governed by electrolyte pH, we next investigated whether the atmospheric open architecture preserves the characteristic electrochemistry of conventional alkaline and acidic Zn–air systems. Rather than comparing their practical performance, we sought to identify the dominant reaction pathways in each electrolyte. Accordingly, 6 M KOH and 3 M ZnCl₂ were arbitrarily selected as representative alkaline and acidic electrolytes, respectively. Under identical conditions, 3 M ZnCl₂ delivered higher operating voltages and discharge currents than 6 M KOH (**Fig. 5a–c**). This behavior is consistent with acidic Zn–air chemistry, where a more positive oxygen-electrode potential and faster ORR kinetics yield higher cell voltages and currents.^{52,53} These results confirm that the electrolyte-in-gas architecture preserves the intrinsic pH-dependent reaction chemistry of Zn–air batteries while providing a distinct interfacial environment.

Ex-situ SEM and XRD analyses further confirmed the pH-dependent reaction pathways. Pristine Zn electrode exhibited a smooth surface, whereas Pt-GDL air electrode displayed its characteristic nanofibrous morphology (**Fig. 5d**). After discharge in 6 M KOH, particulate products formed on the Zn surface, while the Pt-GDL remained morphologically unchanged. By contrast, neither electrode showed discernible morphological changes after discharge in 3 M ZnCl₂. XRD results were consistent with these observations (**Fig. 5e, f**). Zn discharged in 6 M KOH exhibited characteristic ZnO diffraction peaks at $2\theta \approx 31.7^\circ$, 34.4° , and 36.2° , confirming ZnO formation through the typical alkaline Zn-air discharge pathway.⁵⁴ By contrast, no additional crystalline phases were detected after discharge in 3 M ZnCl₂, indicating direct Zn dissolution without solid-product formation, consistent with acidic Zn–air chemistry. In both electrolytes, the Pt-GDL

retained its original diffraction pattern, consistent with ORR proceeding without detectable crystalline solid products.

FT-IR spectroscopy further verified ORR at the positive electrode (**Fig. 5g, h**). After discharge under both alkaline and acidic conditions, broad O–H stretching bands ($3200\text{--}3600\text{ cm}^{-1}$) and H–O–H bending bands at $1630\text{--}1640\text{ cm}^{-1}$, characteristic of surface hydroxyl species and adsorbed H_2O , were consistently observed.⁵⁵⁻⁵⁷ Collectively, these structural and spectroscopic analyses demonstrate that the electrolyte-in-gas architecture preserves the characteristic pH-dependent chemistry of conventional Zn–air batteries, including ZnO formation in alkaline media, direct Zn^{2+} dissolution under acidic conditions, and ORR across a broad pH range.

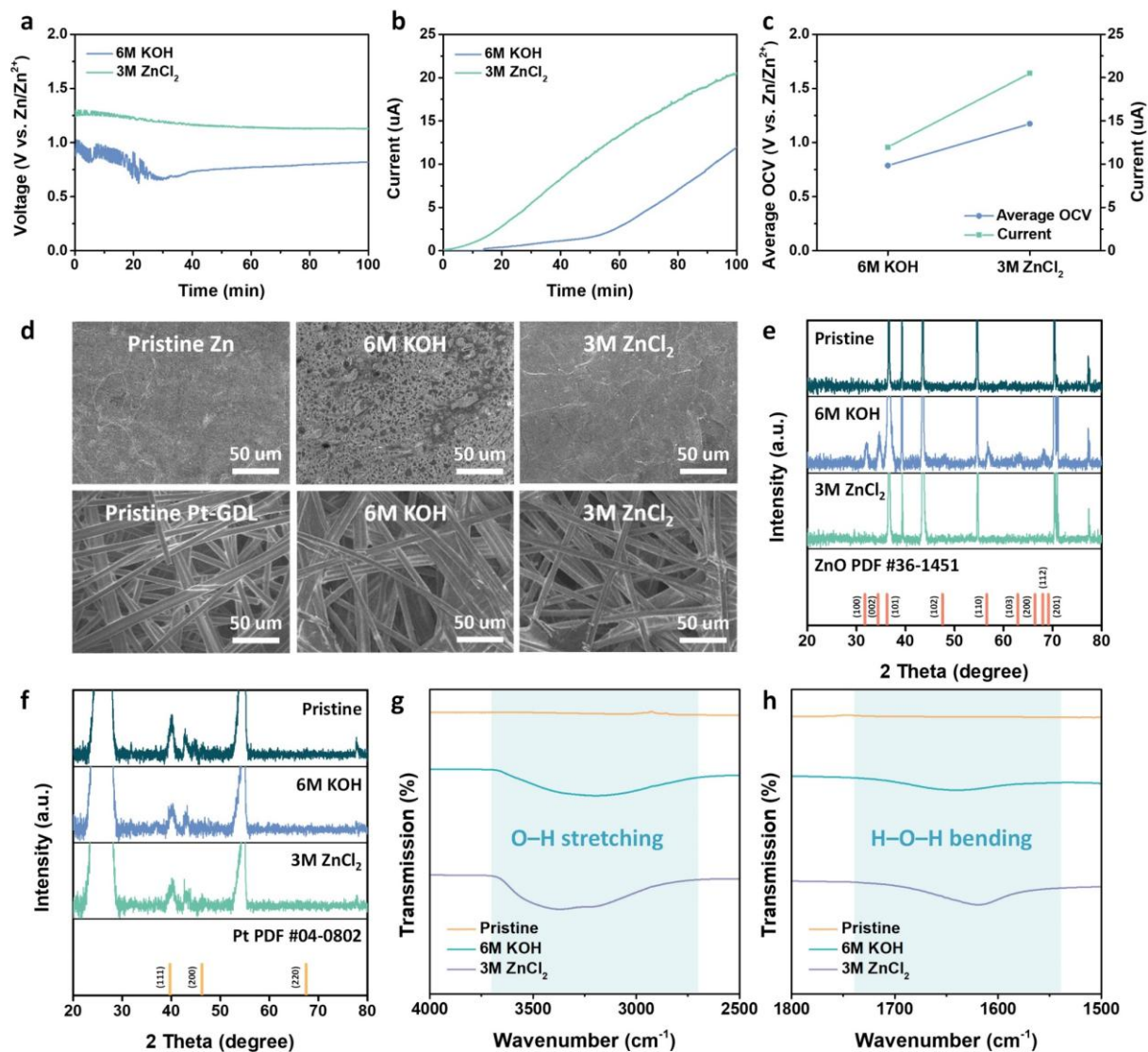


Fig. 5 | Characterisation of Zn-air cell reactions after discharge in different electrolytes. a–c, Electrochemical performance in 6 M KOH and 3M ZnCl_2 aeroelectrolytes: (a) cell voltage profiles, (b) current profiles, and (c) average OCV and current values. **d,** SEM images of Zn metal and Pt-GDL electrodes in the pristine state and after discharge in both electrolytes. **e,f,** XRD patterns of (e) Zn metal and (f) Pt-GDL electrodes in the pristine state and after discharge in both electrolytes.

g,h, FT-IR spectra of Pt-GDL electrodes in the (g) 2500–4000 cm^{-1} and (h) 1500–1800 cm^{-1} regions, measured in the pristine state and after discharge in both electrolytes.

Rechargeability of aerelectrolyte-based atmospheric open Zn–air batteries

Previous study of aerelectrolyte-based Daniell cells was limited to primary (discharge-only) operation as a proof of concept for the atmospheric open electrochemical architecture.³⁰ Here, we extend this concept by demonstrating, for the first time, rechargeable operation of an aerelectrolyte-based atmospheric open Zn–air battery. Electrochemical measurements were conducted using a Zn negative electrode, a Pt-GDL air electrode, and 6 M KOH, with ZnO as the primary discharge product. To directly probe the charging reaction, a ZnO-coated Pt-GDL (ZnO@Pt-GDL) model electrode was used for *ex-situ* and *in-situ* analyses (**Extended Data Fig. 10 and Supplementary Fig. 3**). During charging, oxygen evolution at the Pt-GDL is coupled with electrochemical oxidation of ZnO to soluble zincate species. Linear sweep voltammetry (LSV) exhibited an onset potential of 0.81 V (**Fig. 6a**). Accordingly, constant-voltage measurements produced negligible current at 0.5 V but substantial charging currents at 1 and 2 V, confirming activation of the charging reactions (**Fig. 6b,c**).

The charging reaction was further verified by gas chromatography (GC) and pH measurements. The O₂/N₂ ratio increased from 0.79485 under open-circuit conditions to 1.20217 at a charging voltage above the OER onset potential, confirming oxygen evolution at the air electrode (**Fig. 6d and Supplementary Figs. 4 and 5**). Because OER consumes hydroxide ions, local pH changes were monitored using colorimetric indicators (**Fig. 6e and Supplementary Fig. 6**). The bulk 6 M KOH electrolyte had an initial pH of ~14, which decreased to ~9 after aerosolisation and approached near-neutral values during charging owing to continued OH[−] consumption. These results provide direct evidence of reversible oxygen evolution during charging in the atmospheric open Zn–air battery.

Structural and spectroscopic analyses further confirmed reversible ZnO conversion. The as-prepared ZnO@Pt-GDL electrode exhibited characteristic ZnO diffraction peaks at $2\theta \approx 31.7^\circ$, 34.4° , and 36.2° , which disappeared after charging, confirming electrochemical removal of ZnO (**Fig. 6f**). Consistently, Zn 2p XPS signals assigned to Zn^{2+} (Zn 2p_{3/2} and Zn 2p_{1/2} at 1022.2 and 1045.3 eV, respectively) became undetectable after charging, further supporting complete ZnO decomposition (**Fig. 6g**).⁵⁸ The O 1s spectra likewise showed reversible ZnO evolution (**Fig. 6h**).⁵⁹ After discharge, signals associated with ZnO, surface hydroxyls, and adsorbed H₂O became prominent, whereas the spectrum returned to that of pristine Pt-GDL after charging, demonstrating effective electrode-surface regeneration.

Finally, repeated constant-voltage cycling between 0.2 V (discharge) and 2.0 V (charge) maintained stable voltage and current responses for 250 min (**Fig. 6i**), demonstrating reversible operation and cycling stability. Collectively, the electrochemical, gas, and spectroscopic analyses validate rechargeable atmospheric open Zn–air batteries enabled by the electrolyte-in-gas architecture (**Fig. 6j**). These findings establish a viable rechargeable metal–air battery architecture and provide a broader design framework for gas-involved electrochemical systems.

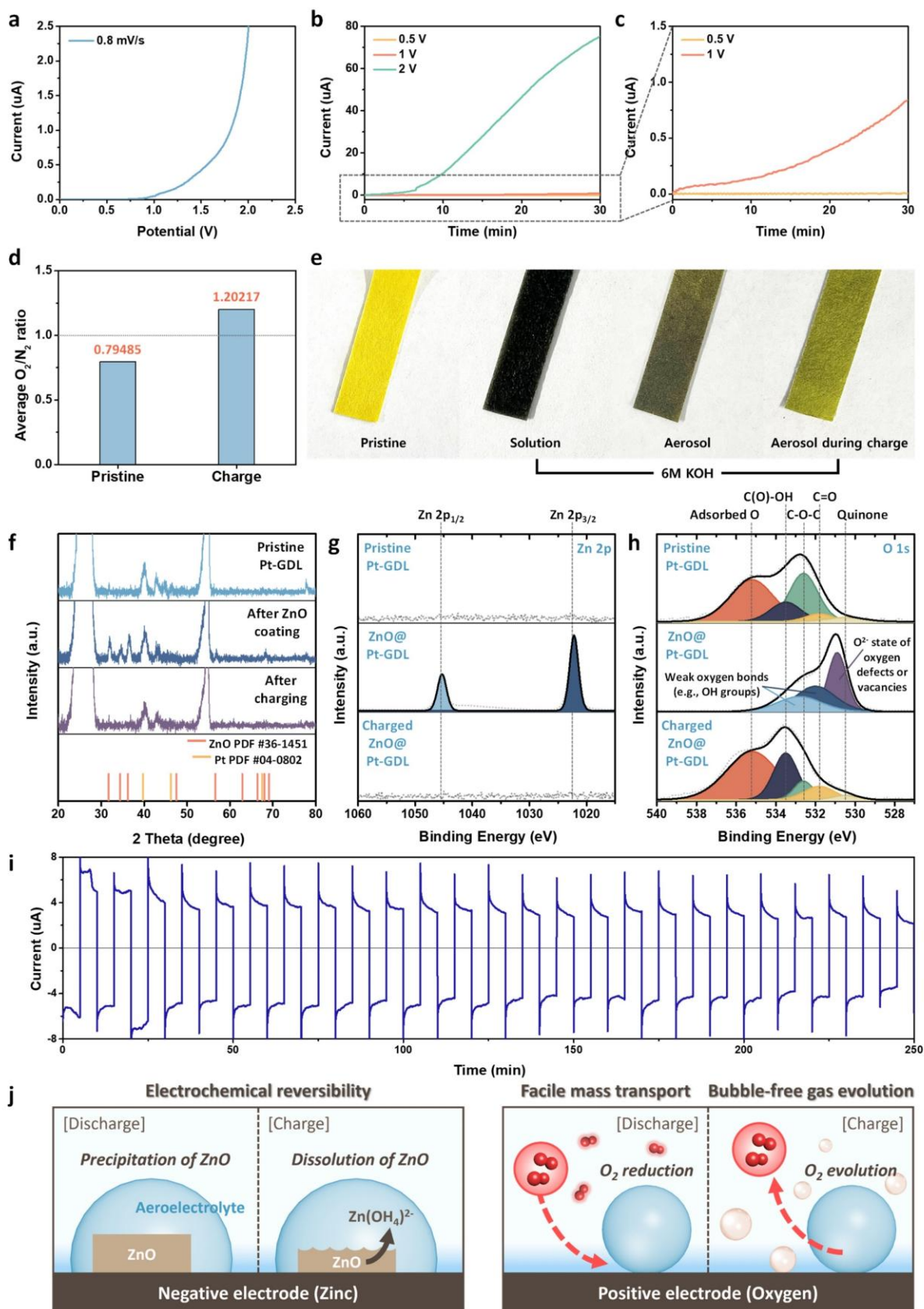


Fig. 6 | Rechargeability of the aerelectrolyte-based alkaline Zn–air battery. **a**, Linear sweep voltammetry (LSV) curve measured at a scan rate of 0.8 mV s^{-1} . **b,c**, Current responses during constant-voltage charging at 0.5, 1.0, and 2.0 V. **d**, Gas chromatography (GC) analysis comparing open-circuit and charged states. **e**, Colorimetric pH analysis using indicator paper for bulk, aerosolized, and electrochemically charged 6 M KOH electrolyte. **f**, XRD patterns of Pt-GDL electrodes in pristine, ZnO-coated and charged states. **g,h**, XPS spectra of (g) Zn 2p and (h) O 1s for Pt-GDL electrodes under the same three conditions. **i**, Cycling performance of the aerelectrolyte-based alkaline Zn–air cell. **j**, Schematic overview of the advantages of rechargeable atmospheric open Zn–air batteries.

CONCLUSIONS

In summary, we introduce an electrolyte-in-gas architecture for atmospheric open metal–air batteries by replacing the conventional bulk electrolyte with continuously supplied aerelectrolyte microdroplets. This architecture spatially decouples gas and ion transport, enabling direct oxygen access to the electrochemical interface while continuously renewing gas–liquid–solid TPBs. A parallel electrode arrangement combined with a porous nanofibrous salt bridge maintains stable ionic conduction without a bulk electrolyte reservoir. Using ZABs as a model system, we established key design principles through electrode engineering, aerelectrolyte transport optimisation, interfacial molecular analysis, and flow visualisation. Aerosolisation was shown to redistribute interfacial ions and modulate local pH relative to the bulk electrolyte. The atmospheric open Zn–air battery also preserved characteristic pH-dependent Zn–O₂ reaction pathways and enabled reversible ZnO redox chemistry, demonstrating rechargeable operation. Beyond improving mass transport and mitigating gas accumulation, the electrolyte-in-gas architecture represents a different electrochemical reaction medium and offers a general framework for rechargeable atmospheric open gas-involved energy-storage technologies.

Methods

Electrode Preparation

Zn metal (Welcos) was employed as the negative electrode. Depending on the experimental requirements, carbon paper (SGL Carbon), Pt-loaded carbon paper (CNL Energy), Ni foam (MTI), Ni mesh (Welcos), and Ni foil (Welcos) was used as the positive electrode. Each electrode was cut to dimensions of 1.8 cm $\times$ 3.6 cm (width $\times$ height) and fixed to a supporting paper substrate with an interelectrode gap of 0.5 cm to ensure consistent alignment during testing.

Salt bridge fabrication via electrospinning

The salt bridge precursor solution was prepared by dissolving 0.09 g potassium nitrate (KNO_3 , 99.0%), 0.5 g polyacrylonitrile (PAN, $M_w = 150,000$), and 0.5 g polyethylene oxide (PEO, $M_v = 100,000$) in 6 mL dimethylformamide (DMF, 99.8%), followed by magnetic stirring at 60 °C for 12 h. For electrospinning, the solution was loaded into a 10 mL Luer-lock syringe mounted on a syringe pump and delivered through a 27-gauge needle at 0.3 mL h^{-1} . A voltage of 16 kV was applied with a tip-to-collector distance of approximately 20 cm. Salt bridge density was controlled by varying the electrospinning time; unless otherwise noted, samples were electrospun for 10 min. The resulting salt bridge morphology was characterised by scanning electron microscopy (SEM; Hitachi).

Electrolyte Preparation and aerosolisation

Zinc chloride (ZnCl_2 , 99.0%) and potassium hydroxide (KOH, 90.0%) were dissolved separately in distilled water to prepare acidic and alkaline aqueous electrolytes, respectively. To generate the aereoelectrolyte, 50 mL of the prepared electrolyte was placed in an aerosol atomiser (DS-A103, DONG SUNG Industry Co., Ltd.) and atomised using compressed air.

Electrochemical measurements

All electrochemical measurements were performed using a potentiostat (SP-200, BioLogic). Open-circuit voltage (OCV) was recorded without applying an external potential or current to determine the equilibrium cell voltage. Zero-voltage current (ZVC) measurements were conducted by applying 0 V between the working and reference electrodes and recording the current required to reduce the cell voltage to zero. The resulting current was mirrored along the x-axis to represent the actual direction of current flow. Linear sweep voltammetry (LSV) was performed from 0 to 2 V at a scan rate of 0.8 mV s^{-1} to evaluate reaction onset and redox kinetics. For constant-voltage (CstV) measurements, fixed potentials of 0.5 V, 1.0 V, and 2.0 V (vs. reference) were applied.

Aereoelectrolyte Characterisation

The dynamic behavior of the aereoelectrolyte was visualised using a high-speed camera (HAS-D71, DITECT), and particle velocity fields were quantitatively analysed using particle image velocimetry (PIV) software (Flownizer 2D, DITECT). Electrode wettability was evaluated by measuring the static contact angle of electrolyte droplets on the electrode surfaces using a contact-angle goniometer (DSA 25, KRÜSS).

Ex-situ Characterisation

Electroplated electrodes were prepared by operating the aerelectrolyte-based cell under either discharge or charge conditions for 2 h. After electrochemical treatment, the nanofiber salt bridge was carefully removed, and the electrodes were washed with distilled water and vacuum-dried. *Ex situ* surface analysis was conducted using X-ray diffraction (XRD; D8 Advance, Bruker) and X-ray photoelectron spectroscopy (XPS; K-Alpha, Thermo Fisher Scientific, UK). Gas chromatography was performed using an HP-PLOT/Q capillary column (Agilent Technologies). The oven, thermal conductivity detector (TCD), and flame ionisation detector (FID) temperatures were set to 150 °C, 200 °C, and 30 °C, respectively. The carrier-gas flow rate was 5.0 mL min⁻¹ and 10 µL of gas was injected per run. Each condition was analysed in triplicate, and O₂/N₂ ratios were calculated from integrated peak areas and averaged. Colorimetric pH analysis was performed using universal indicator paper.

Molecular dynamics simulation

To elucidate the aerosolisation-induced local pH shift and interfacial ion redistribution in 6 M KOH at the molecular level, molecular dynamics (MD) simulations were performed using BIOVIA Materials Studio 2025 (Accelrys Inc.).⁶⁰ All simulations employed the COMPASS III force field, which has been widely used to describe condensed-phase systems containing metal salts, aqueous solutions, and gas phases, providing a reliable description of aqueous systems.⁶¹

Simulation cells were constructed using the Amorphous Cell module in a symmetric slab geometry, with a 6 M KOH aqueous phase centred between gas phases on both sides. Combined with three-dimensional periodic boundary conditions, this configuration generated two gas–liquid interfaces, improving statistical sampling and facilitating interfacial analysis. The liquid phase consisted of 1,000 H₂O molecules and 105 KOH molecules. Since reproducing the actual gas partial pressure

within an aerosol droplet is impractical at the MD simulation scale, the number of gas molecules under each condition (N_2 , O_2 , or air) was adjusted to achieve a gas-phase mass density of 1 g cm^{-3} .^{40,50,51} Applying the same density across all conditions allowed the interfacial effect to be isolated. For the air condition, N_2 and O_2 were mixed at a molar ratio of 8:2 to approximate atmospheric composition. A pure-liquid system without a gas phase served as the control, confirming uniform distributions of OH^- , H_2O , and K^+ in the absence of a gas–liquid interface.

Electrostatic and van der Waals interactions were calculated using Ewald and atom-based summation methods, respectively, with a cutoff distance of $12.5 \text{ \AA}$. initial configurations were energy-minimised, followed by 100 ps of NVT pre-equilibration at 298 K and 2 ns of NVT equilibration. The NVT (canonical) ensemble was adopted instead of NPT to maintain a fixed cell volume and stabilise the two interfaces in the slab geometry. A 20 ns production run was then performed with a 1 fs time step, and temperature was regulated using a Nosé–Hoover thermostat. From the production trajectories, spatial distribution profiles of OH^- , H_2O , K^+ , and gas molecules along the slab-normal direction were computed. The width of the OH^- -rich core and the thickness of the interfacial OH^- depletion layers were then quantitatively evaluated.

Air-Electrolyte transport simulation

A three-dimensional CFD-based particle-tracking model was developed using COMSOL Multiphysics 6.2 to analyse the pressure- and concentration-dependent transport of aereoelectrolyte droplets within the open Zn–air battery reactor. The computational domain reflected the experimental reactor, including the aerosol inlet and outlet geometries. Air was treated as the continuous phase, and a steady-state flow field was first calculated under specified aerosol-

generation pressures. The inlet velocity was determined from the pressure-dependent flow characteristics of the DS-A103 aerosol sprayer.

The airflow was assumed to be incompressible, and the governing equations were expressed as follows:

The flow field was assumed to be incompressible, and the governing equations are as follows.

$$\rho(u \cdot \nabla)u = -\nabla p + \nabla \cdot [\mu(\nabla u + (\nabla u)^T)]$$

Here, ρ is the air density, u is the fluid velocity, p is the pressure, and μ is the dynamic viscosity.

The resulting steady-state flow field was subsequently used as the background velocity field for particle-tracing analysis.

Aeroelectrolyte droplet transport was calculated using the Particle Tracing for Fluid Flow interface. The droplets were treated as dilute discrete liquid particles dispersed in air, with a fixed diameter of 300 nm under all conditions. Droplet density was set according to the solution density at each ZnCl_2 concentration. Particles were emitted from random positions at the aerosol inlet, with initial velocities equal to the local inlet air velocity. Their motion was calculated using a Lagrangian approach according to:

$$\nabla \cdot u = 0$$

$$m_p \frac{dv_p}{dt} = F_D$$

Here, m_p is the particle mass, v_p is the particle velocity, and F_D is the drag force exerted by the continuous airflow.

Two simulation sets were performed. For pressure-dependent simulations, the ZnCl_2 concentration was fixed at 3 M and the aerosol-generation pressure was varied from 0.1 to 0.25 MPa. For concentration-dependent simulations, the pressure was fixed at 0.25 MPa, and the experimentally observed increase in aerosol generation with increasing ZnCl_2 concentration was reflected by increasing the number of emitted particles. The simulations were used to compare droplet trajectories, velocity distributions, internal recirculation behavior, and local particle densities near the electrodes.

Table 1. Parameters used in the particle tracing model

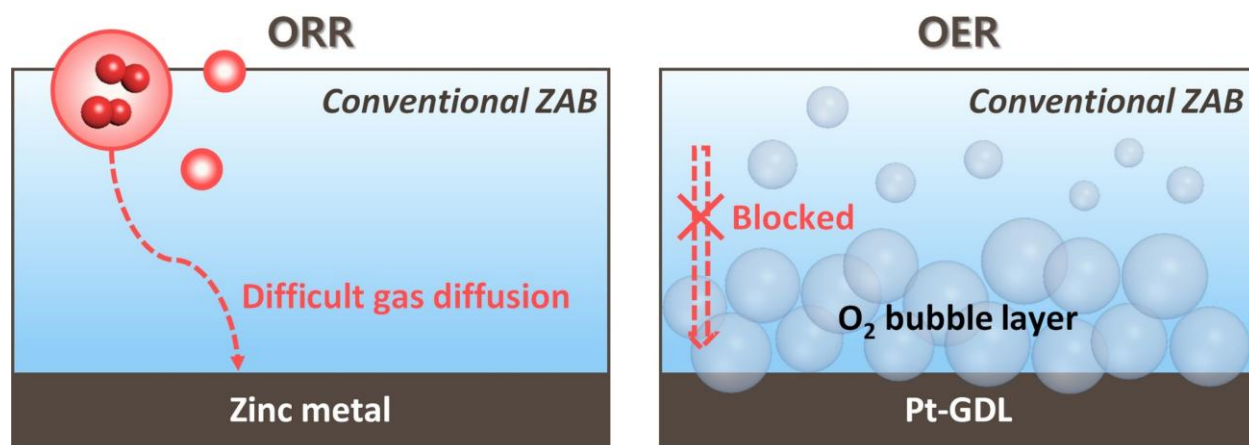
Parameters	Symbol	Value
Reactor length (m)	L	0.1
Reactor depth (m)	D	0.004
Reactor height (m)	H	0.10
Inlet nozzle radius (mm)	R_{nos}	3.175
Inlet nozzle diameter (mm)	d_{nos}	6.35
Outlet open-boundary width (mm)	W_{out}	40
Outlet open-boundary height (mm)	H_{out}	25
Air density (kg m^{-3})	ρ_{air}	1.2
Air dynamic viscosity (Pa s)	μ_{air}	1.8E-5
Droplet diameter (nm)	d_p	300
Inlet particle velocity	$v_{p,0}$	$U_{spray} = U_{in}$

ZnCl ₂ solution density (kg m ⁻³)	ρ_p	1009.85, 1109.22, 1303.48, 1490.59
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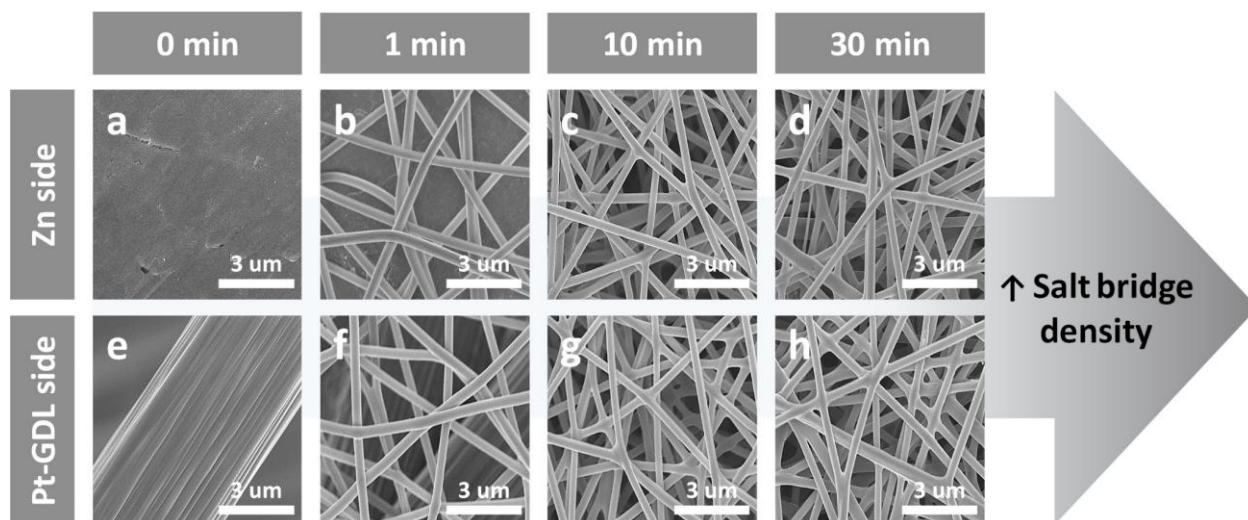
Acknowledgements

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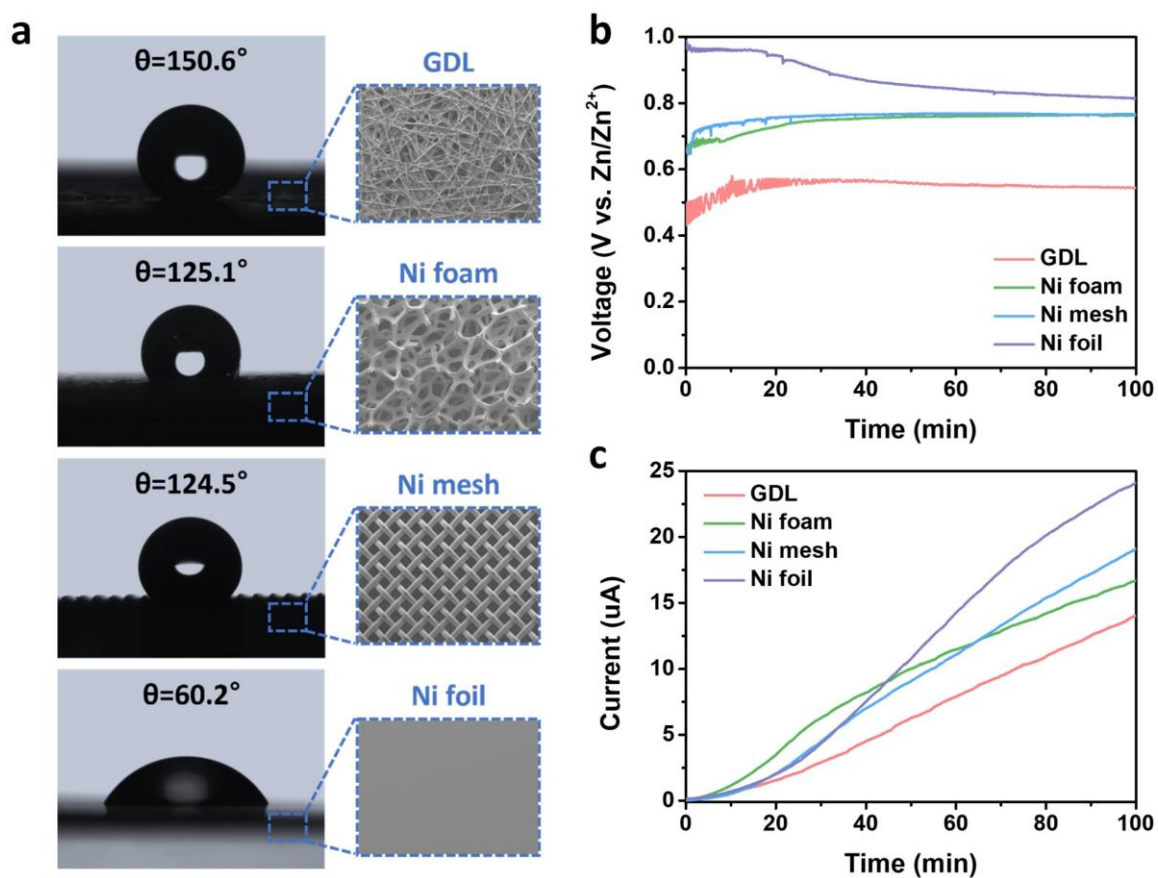
Extended data figures



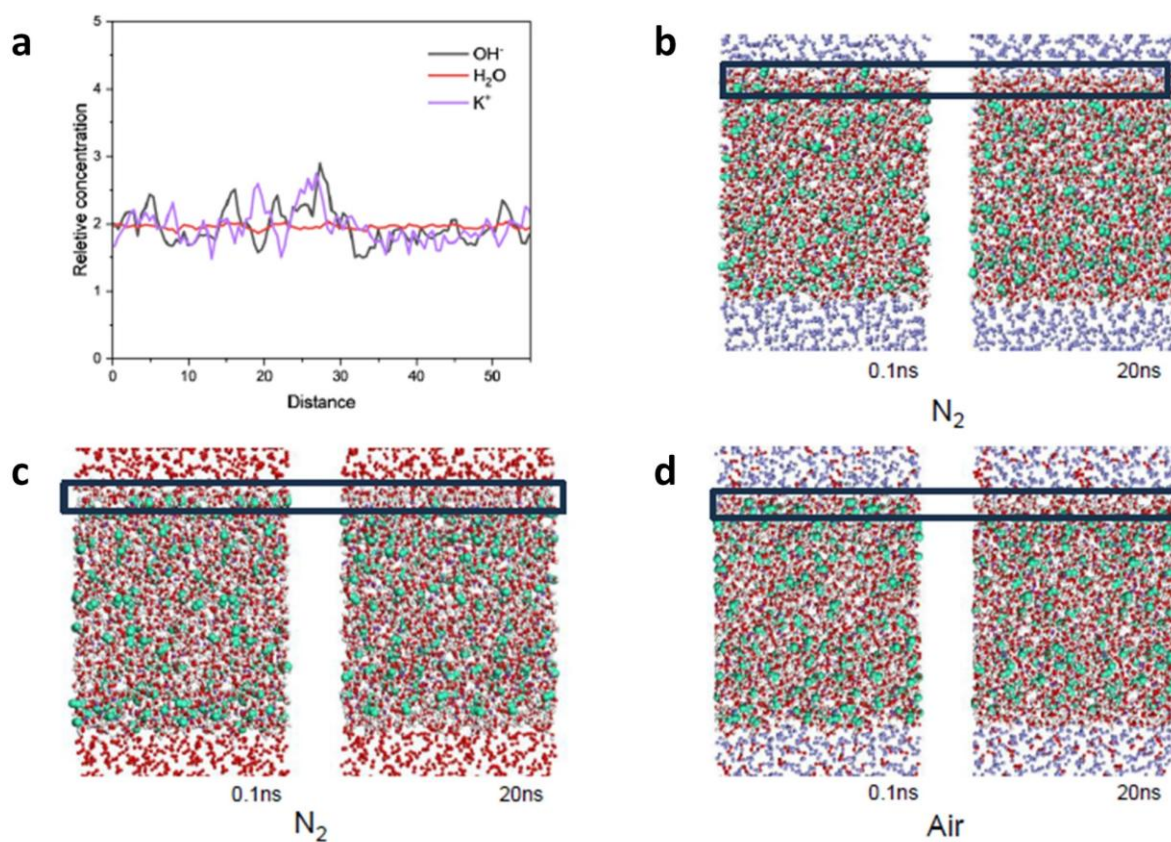
Extended Data Fig. 1 | Schematic illustration of ORR and OER limitations in a conventional Zn-air battery.



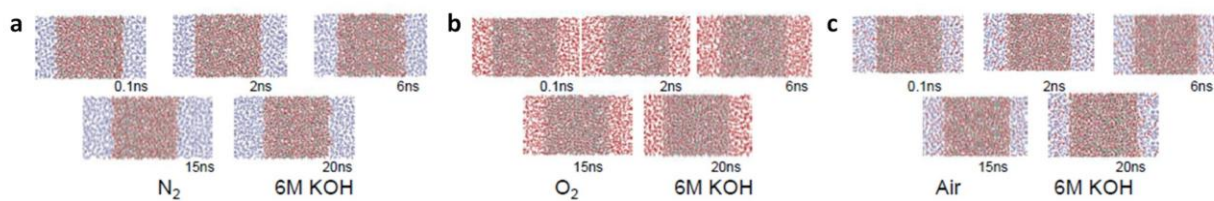
Extended Data Fig. 2 | SEM images of salt-bridge-coated electrodes at various electrospinning times. a–d, Zn side after electrospinning for (a) 0, (b) 1, (c) 10, and (d) 30 min. e–h, Pt-GDL side after electrospinning for (e) 0, (f) 1, (g) 10, and (h) 30 min.



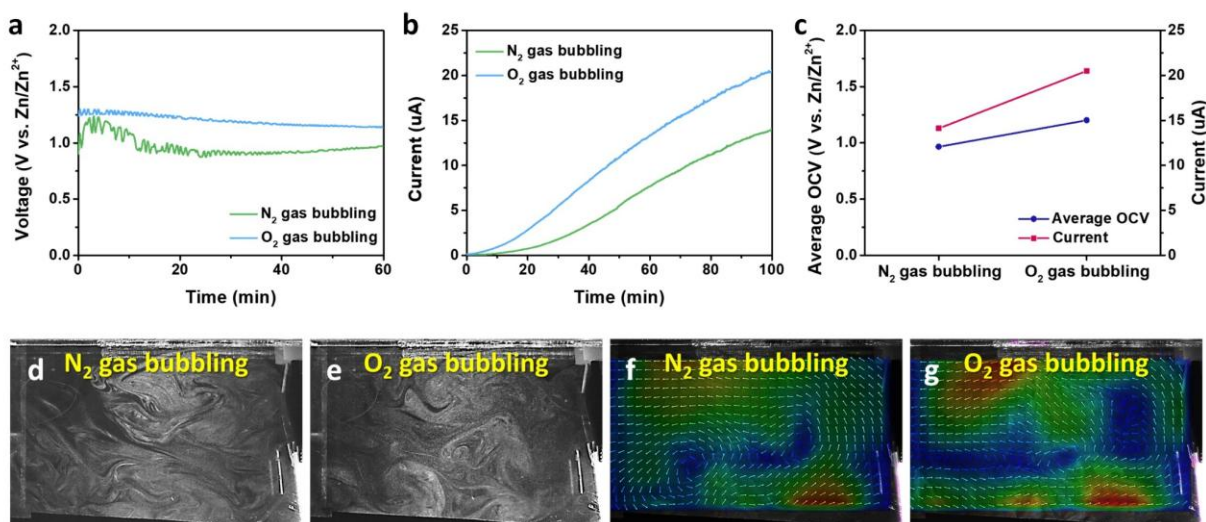
Extended Data Fig. 3 | Electrochemical performance of the aerelectrolyte cell with different electrode configurations. **a**, Contact-angle measurements of the electrolyte on GDL, Ni foam, Ni mesh, and Ni foil electrodes, accompanied by SEM images of the corresponding electrode surfaces. **b,c**, Electrochemical performance with various positive electrodes: (b) cell voltage profiles and (c) current profiles.



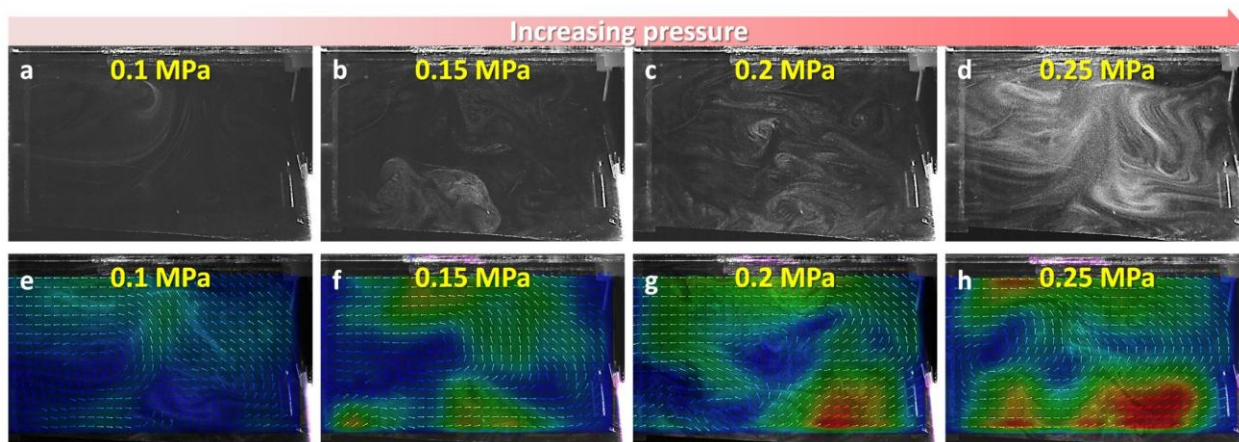
Extended Data Fig. 4 | Relative density profiles and gas-liquid interfacial structures from molecular dynamics simulations. **a**, Relative density profile of the bulk electrolyte. **b–d**, Molecular dynamics snapshots of the gas-liquid interface at 0.1 and 20 ns under (b) N₂, (c) O₂ and (d) air environments.



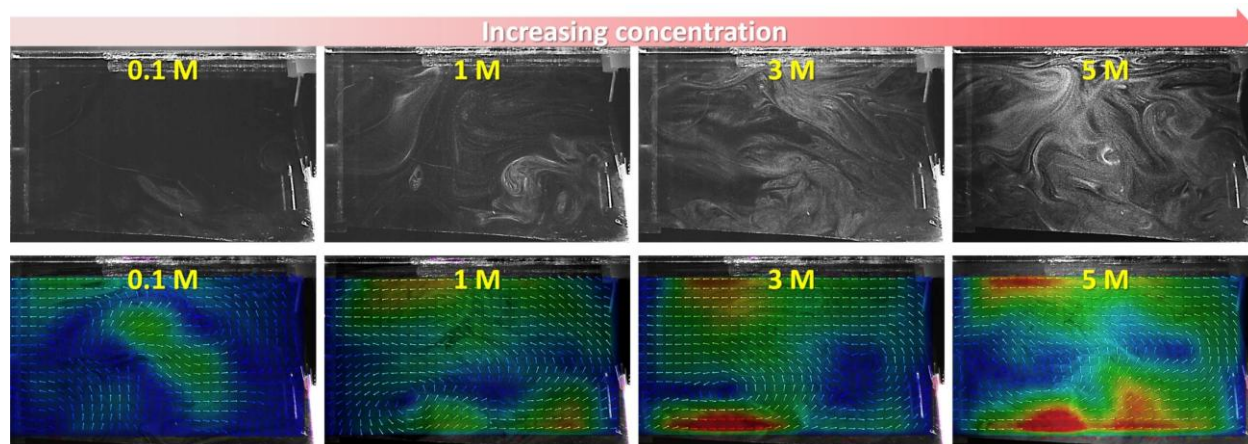
Extended Data Fig. 5 | Time evolution of gas-liquid interfacial structures in molecular dynamics simulations. Molecular dynamics snapshots of the gas-liquid interface under (a) N₂, (b) O₂, and (c) air environments at 0, 2, 6, 15, and 20 ns.



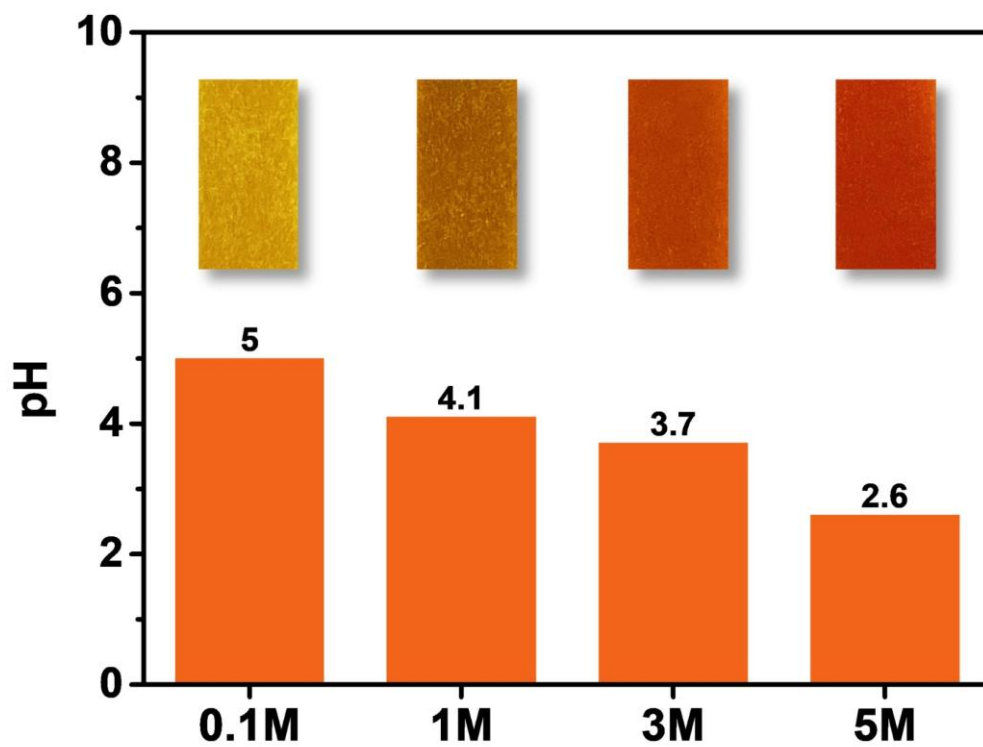
Extended Data Fig. 6 | Effect of dissolved gas species on aeroelectrolyte transport and electrochemical performance. **a-c**, Electrochemical performance at constant pressure (0.25 MPa) and ZnCl_2 concentration (3 M) with different gas-purging conditions: (a) cell voltage profiles, (b) current profiles, and (c) average OCV and current values. **d,e**, High-speed camera images of the aeroelectrolyte for (d) N_2 and (e) O_2 purging. **f,g**, Corresponding flow-rate data for (f) N_2 and (g) O_2 purging.



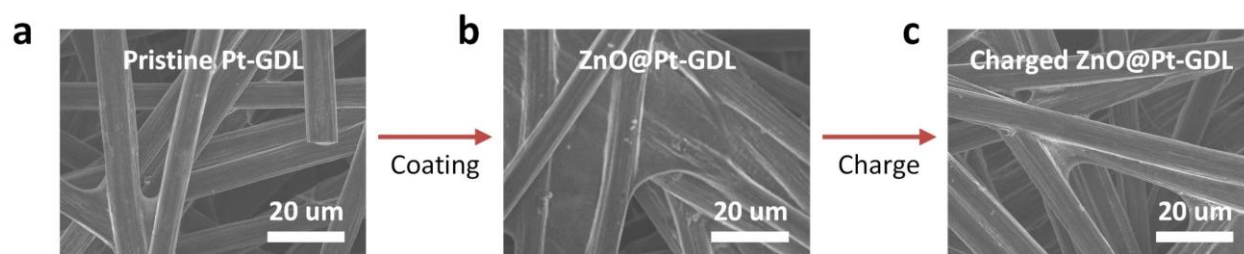
Extended Data Fig. 7 | Effect of aerosol generation pressure on aeroelectrolyte transport. **a–d**, High-speed camera images of the aeroelectrolyte (3 M ZnCl_2 with O_2 purging) generated at pressures of (a) 0.1, (b) 0.15, (c) 0.2, and (d) 0.25 MPa. **e–h**, Corresponding flow-rate data at (e) 0.1, (f) 0.15, (g) 0.2, and (h) 0.25 MPa.



Extended Data Fig. 8 | Effect of ZnCl_2 concentration on aereoelectrolyte transport. a–d, High-speed camera images of the aereoelectrolyte under O_2 purging at 0.25 MPa with ZnCl_2 concentrations of (a) 0.5, (b) 1.0, (c) 3.0, and (d) 5.0 M. e–h, Corresponding flow-rate data for (e) 0.5, (f) 1.0, (g) 3.0, and (h) 5.0 M ZnCl_2 .



Extended Data Fig. 9| pH values and corresponding color changes of indicator paper in ZnCl₂ solutions at 0.1, 1, 3, and 5 M.



Extended Data Fig. 10 | SEM images of Pt-GDL electrodes at different processing stages. a–c, SEM images of the Pt-GDL surface in the (a) pristine state, (b) after ZnO coating, and (c) after electrochemical charging.

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