A Nitrogen Battery Electrode involving Eight-Electron Transfer per Nitrogen for Energy Storage

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Abstract: Redox flow batteries have been discussed as scalable and simple stationary energy storage devices. However, currently developed systems encounter less competitive energy density and high costs, restricting their wider application. There is a lack of appropriate redox chemistry, preferably based on active materials that are abundant in nature and show high solubility in aqueous electrolytes. A nitrogen-centered redox cycle operating between the limiting species ammonia and nitrate via an eight-electron redox reaction stayed practically unnoticed, albeit its ubiquity in biological processes. Ammonia or nitrate are world-scale chemicals with high aqueous solubility, and are then comparatively safe. We demonstrate here the successful implementation of such a nitrogen-based redox cycle between ammonia and nitrate with eight-electron transfer as a catholyte for Zn-based flow batteries, which continuously worked for 12.9 days with 930 charging-discharging cycles. A very competitive energy density of 577 Wh L⁻¹ can be reached, which is well above most reported flow batteries (e.g. 8 times the standard Zn-bromide battery), demonstrating that the nitrogen cycle with eight-electron transfer can offer promising cathodic redox chemistry for safe, affordable, and scalable high-energy-density storage devices.

Redox flow batteries (RFBs) are promising candidates for stationary energy storage devices for modern grids based on intermittent green energy generation.[1] RFBs are unique since electrolyte and electrode are spatially separated, which has the advantages of safety, simplifies scalability and independent tuning of the energy and power output.[2] Besides liquid-liquid RFBs, hybrid-flow batteries combining a multivalent metal (such as Zn, Mg and Al) anode with a redox-active catholyte have attained attention.[3] Until very recently, reported research on electroactive species of the catholyte was rather limited and has been focused mostly on transition metal redox species, halogen, or aqueous soluble organic redox materials.[4] As a mature technology and after a few decades of development, the zinc-bromine RFB has become the reference flow battery for scaled demonstrators, but its industrialization is still hindered by the relatively high cost and low energy density, regarding the low natural abundance of bromine, its high mass, and the low solubility of active materials in the catholyte, respectively.[5] According to the energy density calculation equation (\[E = NCFV/n\]), where \(N\) is the number of electrons transferred in the redox reaction, \(C\) is the concentration of active redox species, \(F\) is the Faraday constant, \(V\) is the voltage, and \(n\) is the number of electrolyte volumes contributing to redox reactions), maximizing \(N, C\) and \(V\) while minimizing \(n\) is beneficial to enhance \(E\).[6]

Until recently, efforts were devoted to exploring novel electroactive materials with high aqueous solubility, yet little attention has been paid to the number of electrons transferred in the redox reaction.[7] Most of the reported cathodic active species such as VO₄³⁻/VO₂⁺, I⁻/I₂⁻, Br⁻/Br₂, Fe(CN)₆₃⁻/Fe(CN)₆₄⁻, Ni(OH)₂/NiOOH, etc. experience single- or two-electron redox reactions while four electrons are involved in the case of Pb redox, (e.g., PbO₂/Pb) (Figure 1a).[8] It is obvious that a higher electron transfer number per unit of active material will significantly reduce the concentration of active material used, thus reducing the costs and increasing the energy density. In this regard, Zn-
Air batteries that integrate a zinc metal anode and a four-electron redox reaction oxygen cathode based on dioxygen (i.e., 2 electrons per oxygen) are worth mentioning. However, the slow kinetics of such a gas-solid-liquid three-phase reaction and carbonate formation when getting in touch with CO$_2$ in air limit practicality.

Another reported competitor is a H$_2$/NaBrO$_3$ flow battery with a six-electron transfer process, but this faces the disadvantages of scarcity of Br and its high mass.

Nitrogen redox chemistry is ubiquitous in the environment and critical to all life, but its applications in electrochemical energy storage are poorly understood. In water, nitrogen is commonly found as nitrate (NO$_3^-$), nitrite (NO$_2^-$) or ammonium ion (NH$_4^+$) with oxidation states of $+V$, $+III$, and $-III$, respectively. At the standard potential of 0.88 V versus normal hydrogen electrode (vs. NHE, pH 0), the transformation between NO$_3^-$ and NH$_4^+$ involves an eight-electron reaction (that is, one electron per 7.7 mass units). This could set the base for a more than competitive active material formulated as a catholyte. For example, such a zinc-nitrogen hybrid flow battery (Zn-N battery, ZNB) has an ideal theoretical energy density of 871 Wh L$^{-1}$ at the solubility limit of KNO$_3$ in the water (38 g/100 mL, 25°C), which is much higher than that of the lead battery, vanadium redox battery, Zn–MnO$_2$, and many others (see Figure 1b). Besides, in terms of the abundance of the active materials, the NO$_3^-$ is a mass chemical, particularly abundant, and can even be retrieved from wastewater by ion exchange in the conventional denitrification process. Very recently, NO$_3^-$ pollutant solutions were used as promising alternatives to dissolved nitrogen and were demonstrated to be easily electro-
converted to valued-added NH$_3$ (NitrRR) under ambient conditions due to the low dissociation energy of the $\equiv$N=O bond (204 kJ mol$^{-1}$) and high absolute solubility of NO$_3^-$ in water.$^{[14]}$ Especially combining the NitrRR-based cathode with metal anode as a galvanic cell for not only NH$_3$ synthesis but electricity production is highly attractive.$^{[15]}$ NH$_3$, on the other hand, is synthesized from N$_2$ in over 200 million tons per year by the traditional Haber-Bosch process and is extensively used in fertilizer production.$^{[15b,16]}$ The conversion of NH$_3$ to nitric acid in the industry is operated by the Ostwald process with noble metals (Pt, Rh) as catalysts under conditions of high temperatures (800–930°C) and pressures (4–15 bar).$^{[17]}$ Although the electrochemical oxidation reaction of NH$_3$ to NO$_2^-$ with high selectivity has not been reported, the ammonia oxidation reaction (AOR) to NO$_2^-$ is thermodynamically favored with appropriate potentials under all pH conditions, as proved by the standard equilibrium potentials in dependence of pH (Pourbaix diagram, Figure 1c).

On the basis of all that knowledge, here an alkaline Zn-based non-flow battery (Zn–Zn$^{2+}$/NO$_3^-$–NH$_3$) is chosen to demonstrate the feasibility of an aqueous nitrogen cycling process for the cathode, a battery which offers a theoretical operating voltage of 1.08 V (Figure 1d, Eq. (1)–(3) (vs. NHE, pH 14)) and the discussed high energy density. Specifically, a divided cell separated by a Nafion membrane is constructed with a Zn metal and a NiRu catalyst-based cathode. The anolyte is composed of 3.0 M KOH solution mixed with saturated Zn(OH)$_2$ while the catholyte consists of 0.5 M NO$_3^-$ and saturated NH$_3$ alkaline solution (3.0 M KOH), both of which are pumped through two half-cells, respectively. We clearly mention that the choice of Nafion for an alkaline cell is suboptimal and accounts for some of the losses reported below, but the choice was done based on availability.

$\text{Anode} : 4\text{Zn} + 16\text{OH}^- \rightarrow 4\text{Zn(OH)}_2^{2-} + 8\text{e}^- \quad E^o = -1.20 \text{ V}$  
$\text{Overall} : 4\text{Zn} + 8\text{OH}^- + 6\text{H}_2\text{O} \rightarrow 4\text{Zn(OH)}_2^{2-} + 4\text{H}_2\text{O} \quad E^o = 1.08 \text{ V}$  

Electrocatalysts enabling high activity and selectivity for NitrRR and AOR are a prerequisite for the cathodic half-cell. In this regard, recent research work has provided insights into the Cu-, Ni-, Co-, and Ru-based electrocatalysts to realize a full eight-electron NitrRR process.$^{[18]}$ For the AOR process, we tried to screen catalysts and found that Ni-based materials show high catalytic activity and selectivity but unfortunately poor stability when exposed to NH$_3$ with a high concentration. Accordingly, a unique NiRu ball-flower structure was designed, where a Ru “pedal armor” protects the catalyst against NH$_3$ erosion and promotes NitrRR catalysis, while the Ni stamen shows outstanding catalytic activity towards the AOR. In addition to a pair of small redox peaks at $\approx 0.4 \text{ V}$ vs. Ag/AgCl attributed to Ni$^{II}$/Ni$^{III}$ redox reaction,$^{[19]}$ the cyclic voltammogram (CV) curve collected in a three-electrode system shows one pair of distinct redox peaks, demonstrating the designed NiRu ball-flower catalyst is quite active in catalyzing the forward and backward transformation of the NO$_3^-/NH_3$ redox couple (Figure 1e). When the catalyst cathode and zinc anode are assembled, the battery delivers a practical discharging voltage of 0.72 V at 2 mA cm$^{-2}$, which stands for an energy density of 577 Wh L$^{-1}$ (Figure 1f). This is 66.2% of the theoretical value and the losses are mostly attributed to the voltage drop sourced from the imperfect engineering, but as a proof-of-principle, it still much higher than most reported battery systems, such as 35–45 Wh L$^{-1}$ for lead battery, $\approx 65$ Wh L$^{-1}$ for Zn–Br$_2$ battery, and 25 Wh L$^{-1}$ for vanadium flow battery (Figure 2d).$^{[18,5a,20]}$

The discharging capability and charging behavior of Zn–Zn$^{2+}$/NO$_3^-$–NH$_3$ battery was initially explored in a non-flow, stationary cell. The battery can maintain the voltage level across the entire NO$_3^-$ concentration range (Figure 1f), and we took 0.5 M NO$_3^-$ as a model concentration to further analyze the battery behavior. In a discharging process, the battery shows an open-circuit potential (OCP) of 1.39 V vs. Zn/Zn$^{2+}$ by the electrochemical workstation test (Figure 2a), which is consistent with the results of the multimeter test (Inset of Figure 2a and Figure S1). The higher-than-theoretical OCP is ascribed to the reduction of few metal oxidized species.$^{[18a]}$ The conversion of NO$_3^-$ to NH$_3$ in the catholyte is paired with the oxidation of Zn at the anode (Figure S2). Two batteries in series can power an electronic calculator working over 50 hours (Figure S3), indicating the practicality of our very small Zn–Zn$^{2+}$/NO$_3^-$–NH$_3$ battery. The highest capacitive contribution (CC) of 97.1 % for NH$_3$ production can be obtained at a high current density of 50 mA cm$^{-2}$ after a 2-hour discharging process, corresponding to an NH$_3$ production of 7.7 mg g$_{cat}^{-1}$ (Figures S4–6). The discharge voltage shows no obvious fading after running over 35 hours at 2 mA cm$^{-2}$ and maintains at around 0.7 V (Figure S5c). With increasing discharge time at 20 mA cm$^{-2}$, the NH$_3$ content in the electrolyte increases correspondingly. After 50-hour of discharge, 0.2 g NH$_3$Cl salt (identified by the XRD pattern) can be collected (Figure S7), corresponding to an average CC of 92.7 % (Figure S2b and Table S2). A good rate performance was also achieved (Figure 2c). At 0.1 mA cm$^{-2}$, the discharge voltage can reach 0.92 V. The voltage recovers to 0.78 V at 0.5 mA cm$^{-2}$ (initial value is 0.83 V) after operation at a high discharge current density of 10 mA cm$^{-2}$.

In a charging process, NO$_3^-$ ($x=\pm 2$, 3) was found in the saturated NH$_3$ alkaline electrolyte, implying the oxidation of NH$_3$ into NO$_3^-$. The highest CC of 98.1 % was obtained at 5 mA cm$^{-2}$ for NiRu electrode-based battery after 2-hour charging (Figures S8–10), and a CC of 93.3 % was maintained even at a high current density of 20 mA cm$^{-2}$. Coulombic Efficiency is also a key performance indicator for rechargeable batteries. More than 90 % Coulombic efficiency can be found at the current density exceeding 5 mA cm$^{-2}$ (Figures 2e and S5b), which is comparable with reported battery systems.$^{[21]}$ At a current density of over 30 mA cm$^{-2}$, the Coulombic efficiency begins to become lower, ascribed to the occurrence of the parasite oxygen

\[ \text{Anode} : \text{Zn} + 2\text{OH}^- + 2\text{e}^- \rightarrow \text{Zn(OH)}_2^{2-} + \text{H}_2\text{O} \]

\[ \text{Overall} : \text{Zn} + \text{H}_2\text{O} \rightarrow \text{Zn(OH)}_2^{2-} + \text{H}_2 \]

\[ E^o = -0.12 \text{ V} \]
Figure 2. Performance of the rechargeable Zn–Zn\textsuperscript{2+} //NO\textsubscript{3}⁻–NH\textsubscript{3} battery. a) Open-circuit voltage for Zn–Zn\textsuperscript{2+} //NO\textsubscript{3}⁻–NH\textsubscript{3} battery when using NiRu ball-flower and pure Ru electrodes for the nitrogen cycle electrocatalysis (Inset: The open circuit voltage of the battery was recorded with a multimeter in the case of disconnection or connection). b) Capacitive contribution during the 50-hour discharging process at 20 mA cm\textsuperscript{-2} (Capacity here is equal to time multiply current density). c) Charge–discharge curves at various current densities from 0.1 mA cm\textsuperscript{-2} to 10 mA cm\textsuperscript{-2}. d) Practical energy density of the Zn–Zn\textsuperscript{2+} //NO\textsubscript{3}⁻–NH\textsubscript{3} hybrid battery compared with that of several kinds of representative batteries (See the practical energy density from Table S5). e) Coulombic efficiency calculated from charge-discharge product (NO\textsubscript{3}⁻ and NH\textsubscript{3} for discharge process, and NO\textsubscript{2}⁻ and NO\textsubscript{3}⁻ for discharge process). f) Capacitive contribution during 240-hour charging process at 20 mA cm\textsuperscript{-2} (The calculated capacity method is the same as discharging process). g) Charge–discharge polarization curves of NiRu- and pure Ru-electrode based batteries and the resultant power density form the discharge polarization curves. h) Long-term charge and discharge cycles of the Zn–Zn\textsuperscript{2+} //NO\textsubscript{3}⁻–NH\textsubscript{3} flow battery at 0.5 mA cm\textsuperscript{-2} (Inset of (h): Long-term charge and discharge cycles of Zn–Zn\textsuperscript{2+} //NO\textsubscript{3}⁻–NH\textsubscript{3} hybrid battery in H-type cell).

evolution reaction (OER) during the charging process. Zn metal deposition on the anode surface was observed after 2-hour charging at 5 mA cm\textsuperscript{-2} and was confirmed by the XRD, SEM and EDS (see detailed discussions in Figures S11–14). The rate performance for charging was also explored (Figure 2c). The loading voltage (1.76 V) at 0.5 mA cm\textsuperscript{-2} can return to the initial value of 1.78 V after charging at a high current density of 10 mA cm\textsuperscript{-2}. The charge-discharge voltage gap at 0.1 mA cm\textsuperscript{-2} is 0.92 V and can be attributed to the overpotential of AOR and NitrRR. In addition, the voltage efficiency of the Zn–N battery was calculated based on charge–discharge curves at various current densities (Table S3). A high voltage efficiency of 47.50 % can be achieved at 0.1 mA cm\textsuperscript{-2}.

For a short-term charging process, nearly half of the oxidation product is NO\textsubscript{3}⁻ (Figure S9a). The full eight-electron transfer process consecutively dominates when the reaction time is longer. As shown in Figures 2f and S15, Table S4, NH\textsubscript{3} finally converts to NO\textsubscript{3} after 10 days of charge reaction at 20 mA cm\textsuperscript{-2}, and the NO\textsubscript{3}⁻ content linearly increases during that period with a concentration in electrolyte reaching 0.4 mol L\textsuperscript{-1}. The corresponding CC for NO\textsubscript{3}⁻ formation is over 90 % after a 10-day charging process, which explicates the charge capacity of the battery to use the full eight-electron-transfer process. In addition, the discharging polarization curve shows the highest power density of 10.0 mW cm\textsuperscript{-2} (Figure 2g), which is superior to that of recently reported Zn–NO\textsubscript{3}⁻ primary batteries.\textsuperscript{[15a, 22]}

The long-term charge-discharge ability of the hybrid Zn–Zn\textsuperscript{2+} //NO\textsubscript{3}⁻–NH\textsubscript{3} battery was evaluated in an H-type cell and a flow cell (Figures 2h, S16 and S17). For the flow cell, the catholyte and anolyte were stored in two external Teflon fluid reservoirs, respectively, and were cycled by peristaltic pumps (Figure S17). The flow battery was repeatedly charged and discharged at 0.5 mA cm\textsuperscript{-2} for 930 cycles with 20 min per cycle period (12.9 days). After initial activation through the first 35 cycles, the battery showed a slight increase in the loading voltage from 1.58 V in the 35th cycle to 1.72 V in the 500th cycle and finally to 1.76 V in the 930th cycle while the discharge voltage around

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0.70 V (Figure 2h), indicating the hybrid Zn–Zn$^{2+}$/$\text{NO}_3^-$/$\text{NH}_3$ RFB is capable of being recharged and has excellent cycle stability of the catalyst. The increasing loading voltage is a known weakness of the zinc anode and can mostly be attributed to this side.

For local monitoring of catalytic mechanism and stability, the material structure and catalytic activity of the NiRu catalyst were further studied in a three-electrode system in detail. The linear sweep voltammetry (LSV) curves show the apparent current density of $\text{NO}_3^-$ reduction and $\text{NH}_3$ oxidation reactions. Especially, a current density of $1 \text{ A cm}^{-2}$ was reached at comparably low potentials of $-0.06 \text{ V}$ for $\text{NO}_3^-$ reduction reaction (Figure 3a), superior to the previous work.\textsuperscript{[11a,18d,22a,23]} The current density of $\text{NO}_3^-$ reduction is similar to that of $\text{NO}_2^-$ reduction while the current density of $\text{NH}_3$ oxidation reaction is lower than that of $\text{NO}_2^-$ oxidation reaction, indicating the activity of catalyst for $\text{NH}_3$ oxidation to $\text{NO}_2^-$ is relatively limited compared to $\text{NO}_2^-$ oxidation to $\text{NO}_3^-$. We then sought to quantify the nitrate/nitrite electrosynthesis from AOR in a 1 M KOH solution containing 0.5 M $\text{NH}_3$ and at a series of potentials with a reaction time of two hours (Figure S18). The total Faradaic efficiency (FE) of the NiRu catalyst for $\text{NO}_x$ ($x = 2, 3$) production gradually declines with a positive shift of applied potentials and reached its highest value of 91.02% at 1.3 V vs. RHE (Figures 3b and S19–S23).

To support the concept of a reversible redox loop of $\text{NH}_3$ and $\text{NO}_x$ ($x = 2, 3$), the NitrRR was also studied by employing the NiRu ball-flower catalyst in 1 M KOH solution containing 0.5 M $\text{NO}_3^-$ (Figures S24–S29). High FE values of 93.6 % for the reduction of $\text{NO}_3^-$ to $\text{NH}_3$ were observed at potentials ranging from $-0.6 \text{ V}$ to 0.2 V vs. RHE (Figure 3c). This demonstrates that the side reactions producing hydrogen or dinitrogen products are much-suppressed during NitrRR processes.\textsuperscript{[24]} On top of that, the slightly strengthened NitrRR performance of Ni–Ru ball-flower structure compared with the mechanical mix of Ru and Ni show that nanometer range electronic interactions play a supportive role (Figures S30–S32): the Ni stamen and Ru petals here integrate to a coupled dyadic system (Figures 3d–f, S33a and S34). As indicated in Figure 3g, lattice distortions are observed at the interface of Ru and

Figure 3. Performance evaluations of electrochemical nitrogen cycle between $\text{NH}_3$ and $\text{NO}_x$ ($x = 2, 3$) and structure characterization of catalysts. a) Linear sweep voltammetry curves of NiRu ball-flower Janus electrocatalysts recorded in various electrolytes show that the nitrogen cycle between $\text{NH}_3$ and $\text{NO}_x$ is possibly achieved. LSV curves were recorded with automatic IR compensation by electrochemical workstation. b) Faradaic efficiency for $\text{NO}_x$ ($x = 2, 3$) synthesis by $\text{NH}_3$ oxidation reaction (AOR) at the applied potential of 1.3 V to 1.8 V vs. RHE (Pink, blue, and green: $\text{NO}_2^-$ Faradaic efficiency. Brown: $\text{NO}_3^-$ Faradaic efficiency). c) Faradaic efficiency for $\text{NH}_3$ synthesis by $\text{NO}_x$ reduction reaction (NitrRR) from the potential of $-0.6 \text{ V}$ to $+0.3 \text{ V}$ vs. RHE. d) TEM images of NiRu ball-flower structure (Inset of (d) is enlargement image). e) STEM and elemental mapping images of NiRu ball-flower sample. f) STEM image and the white line arrow corresponding to the line scan for elemental analysis. g) High-resolution TEM images of NiRu ball-flower nanoparticles. h) XRD patterns of three catalysts. i) Valence electronic signal of three catalysts obtained from XPS measurements. j) Ni and Ru ions concentration in electrolytes detected by ICP after 2-hour AOR for NiRu ball-flower catalyst.
Ni, which indicates the tight junction between the two metals. The results differ from those from the single components or the physical mixture of Ni and Ru, which all show incomplete eight-electron transformation and poor structural stability (Figures S35–S40). High-resolution transmission electron microscopy (TEM) image of NiRu catalyst also shows the interplanar spacing of the Ni (111) plane increases from 0.200 nm or 0.201 nm in pure Ni particles (Figure S37) to 0.217 nm in NiRu catalyst. This enhanced interplanar spacing agrees with the results of X-ray diffraction (XRD) and reflects the hybridization between the Ni and Ru components (Figure 3h).

The electronic interaction between Ni and Ru elements was also followed by X-ray photoelectron spectroscopic (Figure S41). The valence band maximum value and the binding energy of Ni 2p in the mixed metallic NiRu catalyst is close to that of the Ru sample and positive-shifted relative to Ni sample (Figures 3i and S41b). This fits well with the results presented above. It confirms the well-defined generation of charge transfer at the interface between Ni and Ru, as indicated by the fact that the negative peaks (charge depletion) are on the Ru side and the positive peaks (charge accumulation) are on the Ni side from the differential charge density of NiRu structure by the density-functional theory (DFT) computations (Figure S42). Such strong electronic exchange interactions improve the structural and performance stability during electrochemical cycling (Figures S39–S45). For instance, Ni and Ru ions stay almost undetectable in the electrolyte by inductively coupled plasma-mass spectrometry (ICP) when using the NiRu catalyst for two-hour AOR, while approximately 45 ppm of dissolved Ni ions were found for the pure Ni catalyst (Figures 3j and S44, S45). These results indicate that the charge transfer from Ru to Ni inhibits nucleophilic ammonia corrosion.

To further investigate the catalytic mechanism of NiRu Janus electrocatalyst, the electrocatalytic performances of pure Ru catalyst and Ni catalyst for NO$_3^-$ reduction and NH$_3$ oxidation reactions were studied. Based on the poor activity of the Ru catalyst compared to the Ni catalyst, the AOR performance of the NiRu Janus electrocatalyst is mainly derived from the Ni stamen (Figures 3b and S46). In addition, the excellent NitrRR performance mainly originated from the Ru petals in the optimized NiRu catalyst (Figures S47–S51), which can be well illustrated by the inferior or similar performances using pure Ni catalyst and Ru catalyst, respectively, under otherwise unchanged experimental conditions (Figures S47 and S52). Therefore, in the NiRu Janus electrocatalyst, the different roles of Ni stamen and Ru petals promoted outstanding catalytic activity towards the AOR and NitrRR, respectively.

In conclusion, a biomimetic, eight-electron nitrogen cycle was presented as a promising cathode chemistry for the future storage and release of renewable electricity. The concept of the reversible eight-electron conversion between NO$_3^-$ and NH$_3$ was realized with an overall efficiency of up to 91%, based on novel NiRu ball-flower Janus electrocatalyst. The redox couple of NO$_3^-$/NH$_3$ was then coupled with the Zn metal anode to construct a hybrid Zn–N RFB (Zn–Zn$^{2+}$/NO$_3^-$–NH$_3$, RFB), in which the transformation between NO$_3^-$ and NH$_3$ delivered a capacity contribution of around 95%. This hybrid flow battery results in a record high practical energy density of 577 Wh L$^{-1}$, making it stand out from previously reported battery systems. The current experimental set-up realizes already 66.2% of the theoretical capacity, and the differences can be mostly related to the lower cell voltage, which might be caused by the catalytic overpotentials, but is mainly related to inner electric resistances and impedances of our non-optimized demonstrator cell. In addition, if NO$_3^-$ is not completely converted to NH$_3$ with eight-electron transfer during the discharging process, the presence of NO$_2^-$ intermediates could also lead to a loss of energy in the hybrid flow battery.

Importantly, the constructed Zn–N RFB continuously worked for at least 930 charging-discharging cycles (12.9 days), indicating the broad prospects for practical application. We also believe that the performance of the flow battery could be further enhanced by optimizing the setup design, exploiting more efficient electrocatalysts to elevate the eight-electron reaction path, developing tailor-made alkaline separation membranes, and coupling more appropriate anode redox reactions to enlarge the operating voltage. This is the scope of ongoing investigations.

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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data that support the finding of this study are available from the corresponding author upon reasonable request.

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