

Unveiling the Anion Effect of Potassium Salts on Lawsone-Based Anolytes for Aqueous Redox Flow Batteries

Jin Eo

Seoul National University of Science & Technology

Sungmin Park

Seoul National University of Science & Technology

Wonmi Lee

wonmi.lee@kongju.ac.kr

Kongju National University <https://orcid.org/0009-0003-0074-8844>

Yongchai Kwon

Seoul National University of Science & Technology

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Abstract

Aqueous organic redox flow batteries (AORFBs) attract a significant attention for large-scale energy storage systems owing to their intrinsic safety, low cost, and design flexibility. Among various organic redox couples, 2-hydroxy-1,4-naphthoquinone (Lawsone) and ferrocyanide (FeCN) combination exhibits a promising electrochemical performance. However, its practical application is hindered by capacity decay arising from side reactions occurring under strongly alkaline conditions. Although chemical modification of Lawsone proposes to improve cycling stability, such approaches generally require complex synthesis and increase material cost. In this work, supporting electrolyte composition is adjusted to alleviate the strongly alkaline environment without modifying the active material. Specifically, hydroxide is partially replaced with chloride to reduce alkalinity of electrolyte and the effects of potassium salt anions on electrochemical behavior and battery performance of Lawsone anolyte is investigated. Contrary to the initial expectation, conventional 1 M KOH electrolyte exhibits superior electrochemical kinetics, leading to higher capacity utilization and improved cycling stability than the mixed electrolyte (0.8 M KCl + 0.2 M KOH). AORFB employing 1 M KOH retains 72% of its initial capacity after 100 cycles, whereas mixed electrolyte preserves only 42%. These findings indicate that electrochemical performance is governed not only by electrolyte alkalinity but also by the nature of supporting electrolyte anion.

1. Introduction

Aqueous organic redox flow batteries (AORFBs) have emerged as promising candidates for large-scale energy storage systems because of their high safety, flexible scalability, long cycle life, and use of earth-abundant and environmentally benign materials [1–25]. Unlike metal-based flow batteries [26–47], AORFBs employ organic redox-active molecules whose electrochemical properties can be tailored through molecular design, offering opportunities to improve energy density while reducing material cost. Among various organic compounds, quinone-based molecules have attracted considerable attention owing to their fast redox kinetics, reversible electrochemical reactions, and structural diversity [1–12]. In particular, Lawsone (2-hydroxy-1,4-naphthoquinone), which is a naturally occurring quinone extracted from henna plants, has been recognized as a promising anolyte because of its low cost, sustainability, and favorable electrochemical properties [1]. When coupled with the ferri-/ferrocyanide, Lawsone exhibits a relatively high cell voltage and excellent rate capability, making it an attractive candidate for practical AORFB applications.

Most reported Lawsone-based electrolytes employed potassium hydroxide (KOH) as a supporting electrolyte, where deprotonation of the hydroxyl group significantly enhanced the solubility of lawsone while providing rapid electron-transfer kinetics and high electrochemical reversibility. However, strongly alkaline environments also accelerated undesirable side reactions and molecular degradation of quinone species, resulting in progressive capacity decay during long-term cycling. Consequently, considerable efforts have been devoted to improving the stability of lawsone and related quinones through molecular engineering [2–5]. Various functional groups have been introduced into quinone molecules to suppress degradation pathways, enhance solubility, and improve electrochemical stability. Although these

approaches have successfully improved cycling performance, they generally require additional synthesis, purification, and structural characterization, increasing both the complexity and cost of active-material development.

Compared with molecular engineering, electrolyte engineering offers a simpler and more practical strategy because it modifies the electrochemical environment without changing the molecular structure of the active material [6–8]. The composition of supporting electrolytes can influence ionic conductivity, solvation structure, intermolecular interactions, and charge-transfer processes, all of which affect battery performance. Nevertheless, studies on Lawsone-based AORFBs have almost exclusively employed KOH electrolytes, and the influence of supporting electrolyte anions has received little attention.

In this work, to clarify the role of supporting electrolyte anions, various potassium salts were initially screened as candidate supporting electrolytes. Of them, potassium tetraborate ($K_2B_4O_7$), potassium persulfate ($K_2S_2O_8$), and potassium perchlorate ($KClO_4$) were excluded because Lawsone exhibited insufficient solubility in these electrolytes. In contrast to them, potassium sulfate (K_2SO_4) provided adequate solubility but exhibited slower electron-transfer kinetics during cyclic voltammetry measurements than potassium chloride (KCl), making it unsuitable for subsequent full-cell evaluation. Consequently, KOH and a mixed KCl/KOH electrolyte were selected for further investigation. A mixed KCl/KOH electrolyte was initially expected to improve cycling stability by reducing electrolyte alkalinity and suppressing alkaline-induced side reactions. Contrary to this expectation, however, the conventional KOH electrolyte consistently exhibited superior electrochemical performance owing to its faster electron-transfer kinetics. These results demonstrate that the electrochemical behavior of Lawsone cannot be explained solely by electrolyte alkalinity and reveal that the identity of the supporting electrolyte anion plays a critical role in governing charge-transfer behavior and battery performance. This study provides a new insight into the role of supporting electrolyte anions in quinone electrochemistry and highlights electrolyte engineering as an effective strategy for designing high-performance aqueous organic redox flow battery electrolytes.

2. Materials and methods

2.1. Materials

2-Hydroxy-1,4-naphthoquinone (Lawsone, 98%) was obtained from Alfa Aesar. Potassium ferrocyanide trihydrate (ferrocyanide, 99%), potassium hydroxide (KOH, 95%) and potassium chloride (KCl, 99%) were purchased from Samchun Chemicals.

2.2. Electrochemical characterization

Cyclic voltammetry (CV) was conducted to evaluate the redox properties of Lawsone in various conditions of supporting electrolytes using a potentiostat (SP-240, BioLogic). A glassy carbon electrode (GCE), Ag/AgCl electrode, and platinum (Pt) electrode were used for the working electrode, reference electrode, and counter electrode, respectively. The scan rate was 50 mV s^{-1} . Furthermore, the CV of

charged and discharged anolyte and catholyte after the full cell cycling were also measured for analyzing their redox properties.

2.3. Full cell test

For full cell tests with sole KOH salt, 0.25 M potassium ferrocyanide dissolved in 25 mL of 1 M KOH was used for the catholyte and 0.1 M Lawsone dissolved in 20 mL of 1 M KOH was used for the anolyte. To test systems with KCl salt, 0.25 M potassium ferrocyanide dissolved in 25 mL of 1 M KCl was used for the catholyte and 0.1 M Lawsone dissolved in 20 mL of mixture of 0.2 M KOH and 0.8 M KCl was used for the anolyte. A peristaltic pump (BT 100-2 J, Longer Precision Pump Co., Ltd.) with two pump heads (YZ 1515X, Longer Precision Pump Co., Ltd.) and Viton tubes was operated at 30 rpm to transport the electrolytes. Nafion 117 membrane was used to separate the two carbon felt electrodes whose size was $2 \times 3 \text{ cm}^2$. By using a WBCS3000 battery test system from Wonatech, the cut off voltage of 0.2 V ~ 1.6 V was applied and the current density of 40 mA cm^{-2} was used for 100 cycles.

3. Results and discussion

To identify suitable potassium-based supporting electrolytes for Lawsone, the solubility of Lawsone was first evaluated in 0.8 M KOH containing 0.2 M of various potassium salts. As shown in Fig. 1, even 0.1 M lawsone was not completely dissolved in the electrolytes containing $\text{K}_2\text{B}_4\text{O}_7$, $\text{K}_2\text{S}_2\text{O}_8$, or KClO_4 , indicating that these salts adversely affected the solubility of Lawsone. In contrast, 0.1 M Lawsone was fully dissolved in the KOH solution of containing K_2SO_4 or KCl. Therefore, K_2SO_4 and KCl were selected for subsequent half-cell measurements to evaluate their effects on the electrochemical behavior of Lawsone. The dissolution behavior may not exhibit a simple correlation with the size or charge density of the supporting electrolyte anions, suggesting that salt-specific solubility, ion pairing, and solvation effects collectively governed the formation of homogeneous Lawsone electrolytes.

Following the solubility screening, the effects of KCl and K_2SO_4 addition on the electrolyte pH and electrochemical redox properties of Lawsone were investigated using CV measurements. The electrolyte composition was varied by partially replacing KOH with KCl or K_2SO_4 , and the resulting pH values and redox properties were compared with those obtained in 1 M KOH (Fig. 2). The pH of the Lawsone in 1 M KOH electrolyte was 13.94, whereas the pH values decreased to 13.00 and 13.05 in 0.8 M KCl/0.2 M KOH and 0.4 M K_2SO_4 /0.2 M KOH, respectively. When the KOH concentration was further reduced to 0.1 M, the pH of 0.9 M KCl/0.1 M KOH and 0.45 M K_2SO_4 /0.1 M KOH was 11.12 and 6.26, respectively. Thus, partially replacing KOH with KCl or K_2SO_4 generally reduced the alkalinity of the electrolyte.

The redox potential of Lawsone shifted toward more positive values as the alkalinity of the electrolyte decreased. The redox potential increased from -0.707 V in 1 M KOH to -0.695 and -0.672 V in the KCl-containing electrolytes and to -0.693 and -0.667 V in the K_2SO_4 -containing electrolytes. This positive shift can be attributed to the pH-dependent redox behavior of Lawsone, potentially involving changes in its protonation state and solvation environment. Because Lawsone serves as the negative active

material, however, the positive shift in its redox potential is unfavorable for maximizing the full-cell voltage.

The addition of KCl or K_2SO_4 also increased the difference between reduction and oxidation potentials compared with that observed in 1 M KOH. The ΔE_p increased from 0.110 V in 1 M KOH to 0.149 and 0.173 V in the KCl-containing electrolytes and to 0.169 and 0.208 V in the K_2SO_4 -containing electrolytes. Such increased ΔE_p indicates a higher electrochemical polarization and a slower apparent electron-transfer kinetics in the mixed supporting electrolytes. In particular, the K_2SO_4 -containing electrolytes showed larger ΔE_p s than the KCl-containing electrolytes at comparable KOH concentrations, suggesting that the sulfate-containing media were less favorable for the redox kinetics of Lawsone.

The I_{pa}/I_{pc} ratio also deviated further from the ideal value of unity upon the partial replacement of KOH. The ratio decreased from 0.81 in 1 M KOH to 0.68 and 0.48 in the KCl-containing electrolytes and to 0.75 and 0.47 in the K_2SO_4 -containing electrolytes. These changes indicate a general decrease in the apparent redox reversibility of Lawsone as KOH concentration was reduced.

Although addition of KCl or K_2SO_4 resulted in a slower electron transfer and less reversibility, reducing the electrolyte alkalinity by these neutral potassium salts may suppress base-induced side reactions of Lawsone and consequently improve its long-term chemical stability. Therefore, full-cell cycling tests were conducted to determine whether the potential improvement in chemical stability could compensate for the less favorable half-cell electrochemical properties. Among the mixed electrolytes, the KCl-containing electrolyte was selected for subsequent full-cell evaluation because they exhibited smaller ΔE_p s and thus comparatively more favorable apparent electron-transfer kinetics than the corresponding K_2SO_4 -containing electrolyte.

Based on the pH and half-cell CV results, the 0.8 M KCl + 0.2 M KOH electrolyte was determined for full-cell evaluation because this exhibited more favorable redox kinetics than the corresponding K_2SO_4 -containing electrolyte (Scheme 1). Figure 3 compares the electrochemical performance of Lawsone and ferrocyanide included flow batteries employing 1 M KOH or 0.8 M KCl + 0.2 M KOH as the supporting electrolyte for the anolyte. Both cells exhibited nearly identical coulombic efficiencies (99%) and comparable energy efficiencies (79% for 1 M KOH and 78% for 0.8 M KCl + 0.2 M KOH), indicating that partial replacement of KOH with KCl had little influence on the overall efficiency of the cells.

However, a significant difference was observed in capacity utilization and cycling stability. The cell employing 1 M KOH exhibited an initial capacity utilization of 91% and retained 72% after 100 cycles. In contrast, the cell using the mixed KCl and KOH electrolyte showed a similar initial utilization of 92% but retained only 42% after 100 cycles, indicating substantially faster capacity decay during cycling. Therefore, although the mixed electrolyte did not significantly affect the initial electrochemical utilization of Lawsone, it failed to improve the long-term cycling stability.

Initially, it was hypothesized that partially replacing KOH with KCl would reduce the electrolyte alkalinity, thereby suppressing alkaline-induced side reactions of Lawsone and improving cycling stability. To verify this hypothesis, the pHs of both the anolyte and catholyte were measured before and after full-cell operation. The initial pH of the mixed anolyte decreased from 13.9 to 13.0 upon partial replacement of KOH with KCl, while the catholyte pH decreased from 14.0 to 9.9. After 100 cycles, however, they increased to 13.2 and 12.9 for the anolyte and catholyte, respectively, approaching the strongly alkaline conditions observed in the 1 M KOH electrolyte. These results corroborated that the reduced alkalinity of the mixed electrolyte could not be maintained during battery operation. Consequently, the anticipated suppression of alkaline-induced side reactions was not achieved under the present operating conditions.

Considering the CV results shown in Fig. 2, the inferior cycling stability of the mixed electrolyte is likely associated with its less favorable electrochemical kinetics. Compared with 1 M KOH, the KCl-containing electrolyte exhibited a larger peak-to-peak separation and lower apparent reversibility, indicating slower electron-transfer kinetics. Such sluggish charge-transfer behavior may have increased electrochemical polarization and promoted incomplete utilization of the active material during repeated cycling, ultimately leading to accelerated capacity decay. These findings indicate that, although reducing the electrolyte alkalinity was initially expected to improve the chemical stability of Lawsone, the beneficial effect was outweighed by the deterioration in electrochemical kinetics. Therefore, maintaining favorable electron-transfer characteristics provided by hydroxide ions appears to be more critical than simply lowering the electrolyte pH for achieving stable cycling performance in Lawsone-based AORFBs.

To further investigate the origin of the capacity decay, CV curves for both the anolyte and catholyte before and after the full-cell cycling tests are measured (Fig. 4). The CV curve profiles of the lawsone anolyte employing 1 M KOH and 0.8 M KCl + 0.2 M KOH are compared in Fig. 4(a) and 4(c), respectively. After 100 cycles, the redox peaks of Lawsone in the mixed electrolyte of KCl and KOH was better preserved than those in the 1 M KOH electrolyte, suggesting that partial replacement of KOH with KCl may alleviate the degradation of Lawsone to some extent by reducing the initial electrolyte alkalinity. This observation is consistent with the expectation that a less alkaline environment can suppress alkaline-induced side reactions of quinone-based active materials.

In contrast, the opposite trend was observed for the ferrocyanide catholyte. As shown in Fig. 4(b) and 4(d), the ferrocyanide redox peaks were better maintained when the anolyte employed 1 M KOH, whereas a more pronounced decrease in peak current was observed after cycling when the mixed electrolyte of KCl and KOH was used. Combined with the pH measurements, these results corroborated that the catholyte in the mixed-electrolyte cell gradually became more alkaline during cycling, which may accelerate undesirable side reactions of the ferrocyanide electrolyte, reducing its electrochemical activity.

These findings indicate that simply lowering the electrolyte alkalinity is insufficient to improve the cycling stability of Lawsone-based AORFBs. Instead, maintaining adequate hydroxide concentration for favorable electron-transfer kinetics while suppressing alkaline-induced side reactions should be

considered simultaneously. Therefore, future electrolyte engineering should focus not only on reducing the electrolyte pH but also on optimizing the supporting electrolyte composition to balance electrochemical kinetics and chemical stability.

4. Conclusion

This study systematically investigated the influence of supporting electrolyte anions on the electrochemical behavior of Lawsone in aqueous organic redox flow batteries by partially replacing KOH with potassium salts. Among the potassium salts examined, K_2SO_4 and KCl were determined for electrochemical evaluation. Their CV curves revealed that partially replacing KOH with KCl or K_2SO_4 shifted the redox potential to more positive values, increased the peak-to-peak separation, and decreased the apparent reversibility of Lawsone, indicating slower electron-transfer kinetics than in the conventional KOH electrolyte. Full-cell cycling tests further demonstrated that the mixed electrolyte of KCl and KOH exhibited significantly poorer capacity retention despite its lower initial alkalinity. Although the reduced alkalinity partially preserved the electrochemical activity of Lawsone after cycling, it did not improve the overall cycling stability because the beneficial effect was offset by the deterioration in electron-transfer kinetics and the degradation of the catholyte. These results demonstrate that simply lowering the electrolyte pH is insufficient to improve the cycling stability of Lawsone-based AORFBs. Instead, future electrolyte engineering should focus on developing supporting electrolyte compositions that suppress alkaline-induced side reactions while preserving the favorable electron-transfer kinetics provided by hydroxide ions. Such an approach will provide a more rational strategy for designing high-performance Lawsone-based aqueous organic redox flow batteries.

Declarations

Competing interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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References

1. W. Lee, A. Konovalova, E. Tsoy, G. Park, D. Henkensmeier, Y. Kwon, Alkaline naphthoquinone-based redox flow batteries with a crosslinked sulfonated polyphenylsulfone membrane, *Int. J. Energy Res.* **46**, 12988-13002 (2022). <https://doi.org/10.1002/er.8079>

2. D. G. Kwabi, K. Lin, Y. Ji, E. F. Kerr, M. A. Goulet, D. De Porcellinis, D. P. Tabor, D. A. Pollack, A. Aspuru-Guzik, R. G. Gordon, M. J. Aziz, Alkaline quinone flow battery with long lifetime at pH 12. *Joule* **2**, 1894-1906 (2018). <https://doi.org/10.1016/j.joule.2018.07.005>
3. L. Tong, M. A. Goulet, D. P. Tabor, E. F. Kerr, D. De Porcellinis, E. M. Fell, A. Aspuru-Guzik, R. G. Gordon, M. J. Aziz, Molecular engineering of an alkaline naphthoquinone flow battery. *ACS Energy Lett.* **4**, 1880-1887 (2019). <https://doi.org/10.1021/acsenenergylett.9b01321>
4. S. Huang, H. Zhang, M. Salla, J. Zhuang, Y. Zhi, X. Wang, Q. Wang, Molecular engineering of dihydroxyanthraquinone-based electrolytes for high-capacity aqueous organic redox flow batteries. *Nat. Commun.* **13**, 4746 (2022). <https://doi.org/10.1038/s41467-022-32424-8>
5. C. Wang, Z. Yang, Y. Wang, P. Zhao, W. Yan, G. Zhu, L. Ma, B. Yu, L. Wang, G. Li, J. Liu, Z. Jin, High-performance alkaline organic redox flow batteries based on 2-hydroxy-3-carboxy-1, 4-naphthoquinone. *ACS Energy Lett.* **3**, 2404-2409 (2018). <https://doi.org/10.1021/acsenenergylett.8b01296>
6. W. Lee, K. I. Shim, G. Park, J. W. Han, Y. Kwon, Rational design of composite supporting electrolyte required for achieving high performance aqueous organic redox flow battery. *Chem. Eng. J.* **464**, 142661 (2023). <https://doi.org/10.1016/j.cej.2023.142661>
7. W. Lee, G. Park, Y. Kwon, Alkaline aqueous organic redox flow batteries of high energy and power densities using mixed naphthoquinone derivatives. *Chem. Eng. J.* **386**, 123985 (2020). <https://doi.org/10.1016/j.cej.2019.123985>
8. G. Park, W. Lee, M. Shin, Y. Kwon, Stable redox flow batteries through suppression of side reactions with lawsone anolyte and iodine catholyte. *Appl. Energy* **409**, 127490 (2026). <https://doi.org/10.1016/j.apenergy.2026.127490>
9. W. Lee, G. Park, D. Schröder, Y. Kwon, Performance enhancement of alkaline organic redox flow battery using catalyst including titanium oxide and Ketjenblack. *Korean J. Chem. Eng.* **39**, 1624-1631 (2022). <https://doi.org/10.1007/s11814-021-1040-9>
10. W. Lee, G. Park, Y. Kwon, Cost, performance, and sustainability of redox flow batteries using 4, 5-Dihydroxy-1, 3-benzenedisulfonic acid and vanadium enhanced by cross compensation of the activation process. *ACS Sustain. Chem. Eng.* **11**, 12458-12466 (2023). <https://doi.org/10.1021/acssuschemeng.3c03316>
11. W. Lee, G. Park, Y. Kim, D. Chang, Y. Kwon, Nine watt-level aqueous organic redox flow battery stack using anthraquinone and vanadium as redox couple. *Chem. Eng. J.* **398**, 125610 (2020). <https://doi.org/10.1016/j.cej.2020.125610>
12. W. Lee, A. Permatasari, Y. Kwon, Neutral pH aqueous redox flow batteries using an anthraquinone-ferrocyanide redox couple. *J. Mater. Chem. C* **8**, 5727-5731 (2020). <https://doi.org/10.1039/d0tc00640h>
13. D. Emmel, S. Kunz, N. Blume, Y. Kwon, T. Turek, C. Minke, D. Schröder, Benchmarking organic active materials for aqueous redox flow batteries in terms of lifetime and cost. *Nat. Commun.* **14**, 6672 (2023). <https://doi.org/10.1038/s41467-023-42450-9>

14. W. Lee, G. Park, M. Shin, D. Emmel, D. Schröder, Y. Kwon, Challenges and advances in redox flow batteries utilizing sustainable and cost-effective non-vanadium active materials. *J. Mater. Chem. A* **13**, 15491-15516 (2025). <https://doi.org/10.1039/d5ta00113g>
15. S. Jin, E. M. Fell, L. Vina-Lopez, Y. Jing, P. W. Michalak, R. G. Gordon, M. J. Aziz, Near neutral pH redox flow battery with low permeability and long-lifetime phosphonated viologen active species. *Adv. Energy Mater.* **10**, 2000100 (2020). <https://doi.org/10.1002/aenm.202000100>
16. H. Li, M. Wen, W. Dong, Y. Duan, F. Hu, J. Yang, Z. Jiang, H. Fan, B. Hu, R. Mahalingam, J. Song, Viologen Derivatives in Aqueous Organic Redox Flow Batteries: Progress and Perspectives. *Adv. Mater.* **38**, e14004 (2026). <https://doi.org/10.1002/adma.202514004>
17. T. Yin, J. Duanmu, L. Liu, Viologen-based aqueous organic redox flow batteries: materials synthesis, properties, and cell performance. *J. Mater. Chem. A* **12**, 15519-15540 (2024). <https://doi.org/10.1039/d4ta00753k>
18. T. Liu, X. Wei, Z. Nie, V. Sprenkle, W. Wang, A total organic aqueous redox flow battery employing a low cost and sustainable methyl viologen anolyte and 4-HO-TEMPO catholyte. *Adv. Energy Mater.* **6**, 1501449 (2016). <https://doi.org/10.1002/aenm.201501449>
19. H. Li, H. Fan, B. Hu, L. Hu, G. Chang, J. Song, Spatial structure regulation: a rod-shaped viologen enables long lifetime in aqueous redox flow batteries. *Angew. Chem.* **133**, 27177-27183 (2021). <https://doi.org/10.1002/ange.202110010>
20. X. L. Lv, P. T. Sullivan, W. Li, H. C. Fu, R. Jacobs, C. J. Chen, D. Morgan, S. Jin, D. Feng, Modular dimerization of organic radicals for stable and dense flow battery catholyte. *Nat. Energy* **8**, 1109-1118 (2023). <https://doi.org/10.1038/s41560-023-01320-w>
21. P. T. Sullivan, H. Liu, X. L. Lv, S. Jin, W. Li, D. Feng, Viologen hydrothermal synthesis and structure–property relationships for redox flow battery optimization. *Adv. Energy Mater.* **13**, 2203919 (2023). <https://doi.org/10.1002/aenm.202203919>
22. X. L. Lv, P. Sullivan, H. C. Fu, X. Hu, H. Liu, S. Jin, W. Li, D. Feng, Dextrosil-viologen: a robust and sustainable anolyte for aqueous organic redox flow batteries. *ACS Energy Lett.* **7**, 2428-2434 (2022). <https://doi.org/10.1021/acsenergylett.2c01198>
23. K. Lin, R. Gómez-Bombarelli, E. S. Beh, L. Tong, Q. Chen, A. Valle, A. Aspuru-Guzik, M. Aziz, R. G. Gordon, A redox-flow battery with an alloxazine-based organic electrolyte. *Nat. Energy* **1**, 16102 (2016). <https://doi.org/10.1038/nenergy.2016.102>
24. W. Lee, B. W. Kwon, Y. Kwon, Effect of carboxylic acid-doped carbon nanotube catalyst on the performance of aqueous organic redox flow battery using the modified alloxazine and ferrocyanide redox couple. *ACS Appl. Mater. Interfaces* **10**, 36882-36891 (2018). <https://doi.org/10.1021/acsami.8b10952>
25. A. Aspuru-Guzik, M. J. Aziz, R. G. Gordon, A redox-flow battery with an alloxazine-based organic electrolyte. *Cell* **99**, 1-15 (2016). <https://doi.org/10.1038/NENERGY.2016.102>
26. M. Shin, Y. Oh, Y. Kwon, Adenosine-Derivative Functionalized Carbon Nanotubes Considered as Catalysts for Vanadium Flow Batteries, *Korean J. Chem. Eng.* **41**, 3821-3830 (2024).

<https://doi.org/10.1007/s11814-024-00324-w>

27. M. Ulaganathan, V. Aravindan, Q. Yan, S. Madhavi, M. Skyllas-Kazacos, T. M. Lim, Recent advancements in all-vanadium redox flow batteries. *Adv. Mater. Interfaces* **3**, 1500309 (2016).
<https://doi.org/10.1002/admi.201500309>
28. J. Jang, M. Shin, Y. Kwon, C. Jo, Carbon cloth modified by direct growth of nitrogen-doped carbon nanofibers and its utilization as electrode for zero gap flow batteries. *Chem. Eng. J.* **481**, 148644 (2024). <https://doi.org/10.1016/j.cej.2024.148644>
29. G. Park, Y. Lim, K. Hyun, Y. Kwon, Rapid preparation of desirable vanadium electrolyte using ascorbic acid as a reducing agent in vanadium redox flow batteries. *J. Power Sources* **589**, 233770 (2024).
<https://doi.org/10.1016/j.jpowsour.2023.233770>
30. H. Lim, M. Shin, Y. Kwon, Vanadium redox flow batteries including carbon catalysts derived from low-density polyethylene and polyurethane. *Korean J. Chem. Eng.* **40**, 3087-3095 (2023).
<https://doi.org/10.1007/s11814-023-1576-y>
31. Z. Huang, A. Mu, L. Wu, B. Yang, Y. Qian, J. Wang, Comprehensive analysis of critical issues in all-vanadium redox flow battery. *ACS Sustain. Chem. Eng.* **10**, 7786-7810 (2022).
<https://doi.org/10.1021/acssuschemeng.2c01372>
32. Y. Lim, M. Shin, Y. Kwon, Economical preparation of vanadium electrolytes via ascorbic acid-driven reduction of vanadium pentoxide. *Chem. Eng. J.* **521**, 166775 (2025).
<https://doi.org/10.1016/j.cej.2025.166775>
33. T. T. Bui, S. Park, M. Shin, M. M. Ikhsan, Y. Kwon, D. Henkensmeier, Sulfonated polybenzimidazole membranes: how sulfonation affects properties, stability, and performance in vanadium redox flow batteries. *Adv. Energy Mater.* **15**, 2500440 (2025). <https://doi.org/10.1002/aenm.202500440>
34. H. Chen, X. Zhang, S. Wu, F. Chen, J. Xu, A comparative study of iron-vanadium and all-vanadium flow battery for large scale energy storage. *Chem. Eng. J.* **429**, 132403 (2022).
<https://doi.org/10.1016/j.cej.2021.132403>
35. H. Lim, M. Shin, Y. Kwon, Vanadium redox flow batteries including carbon catalysts derived from low-density polyethylene and polyurethane. *Korean J. Chem. Eng.* **40**, 3087-3095 (2023).
<https://doi.org/10.1007/s11814-023-1576-y>
36. K. Hyun, M. Shin, Y. Kwon, Performance evaluation of zero-gap vanadium redox flow battery using composite electrode consisting of graphite and buckypaper. *Korean J. Chem. Eng.* **39**, 3315-3322 (2022). <https://doi.org/10.1007/s11814-022-1262-5>
37. Z. Yuan, Y. Yin, C. Xie, H. Zhang, Y. Yao, X. Li, Advanced materials for zinc-based flow battery: development and challenge. *Adv. Mater.* **31**, 1902025 (2019).
<https://doi.org/10.1002/adma.201902025>
38. Y. Oh, M. Shin, Y. Kwon, Aqueous all-iron flow batteries using surface-modified carbon electrodes improved via electrochemical exfoliation. *J. Mater. Chem. A* **13**, 37244-37258 (2025).
<https://doi.org/10.1039/d5ta05698e>

39. J. Lee, S. Lim, Y. Kwon, Fabrication of Flexible Nickel Hydroxide Electrode for Nickel–Zinc Battery. *Korean J. Chem. Eng.* **42**, 3303-3311 (2025). <https://doi.org/10.1007/s11814-025-00575-1>
40. L. Guo, H. Guo, H. Huang, S. Tao, Y. Cheng, Inhibition of zinc dendrites in zinc-based flow batteries. *Front. Chem.* **8**, 557 (2020). <https://doi.org/10.3389/fchem.2020.00557>
41. S. Park, M. Shin, U. Kunz, Y. Kwon, The performance of all iron-based redox flow batteries enhanced by carbon nanotube catalysts. *Korean J. Chem. Eng.* **41**, 2441-2448 (2024). <https://doi.org/10.1007/s11814-024-00200-7>
42. S. Kim, M. Shin, S. T. Oh, D. H. Kim, Y. Kwon, Aqueous flow battery using iron and oxygen as redox couple and cobalt (triisopropanolamine) as redox mediator. *J. Mater. Sci. Technol.* **237**, 145-154 (2025). <https://doi.org/10.1016/j.jmst.2025.03.024>
43. Q. Wang, W. A. Daoud, Reaction kinetics of cerium on glassy carbon with in situ electrochemical treatment for cerium-based flow battery. *Electrochim. Acta* **315**, 1-8 (2019). <https://doi.org/10.1016/j.electacta.2019.05.083>
44. M. Shin, S. Park, K. Hyun, Y. Kwon, Spectroscopic and electrochemical analyses elucidating capacity degradation mechanism of iron-ligand complex and air in all iron aqueous redox flow batteries. *Chem. Eng. J.* **471**, 144682 (2023). <https://doi.org/10.1016/j.cej.2023.144682>
45. H. Lim, M. Shin, C. Noh, E. Koo, Y. Kwon, K. Y. Chung, Performance evaluation of aqueous all iron redox flow batteries using heat treated graphite felt electrode. *Korean J. Chem. Eng.* **39**, 3146-3154 (2022). <https://doi.org/10.1007/s11814-022-1195-z>
46. P. K. Leung, M. R. Mohamed, A. A. Shah, Q. Xu, M. B. Conde-Duran, A mixed acid based vanadium–cerium redox flow battery with a zero-gap serpentine architecture. *J. Power Sources* **274**, 651-658 (2015). <https://doi.org/10.1016/j.jpowsour.2014.10.034>
47. K. Amini, M. D. Pritzker, In situ polarization study of zinc–cerium redox flow batteries. *J. Power Sources* **471**, 228463 (2020). <https://doi.org/10.1016/j.jpowsour.2020.228463>

Scheme 1

Scheme 1 is available in the supplementary files section.

Figures



Figure 1

A photo showing the precipitation of 0.1 M Lawsone in 0.8 M KOH containing 0.2 M i) $K_2B_4O_7$, ii) $K_2S_2O_8$, or iii) $KClO_4$.

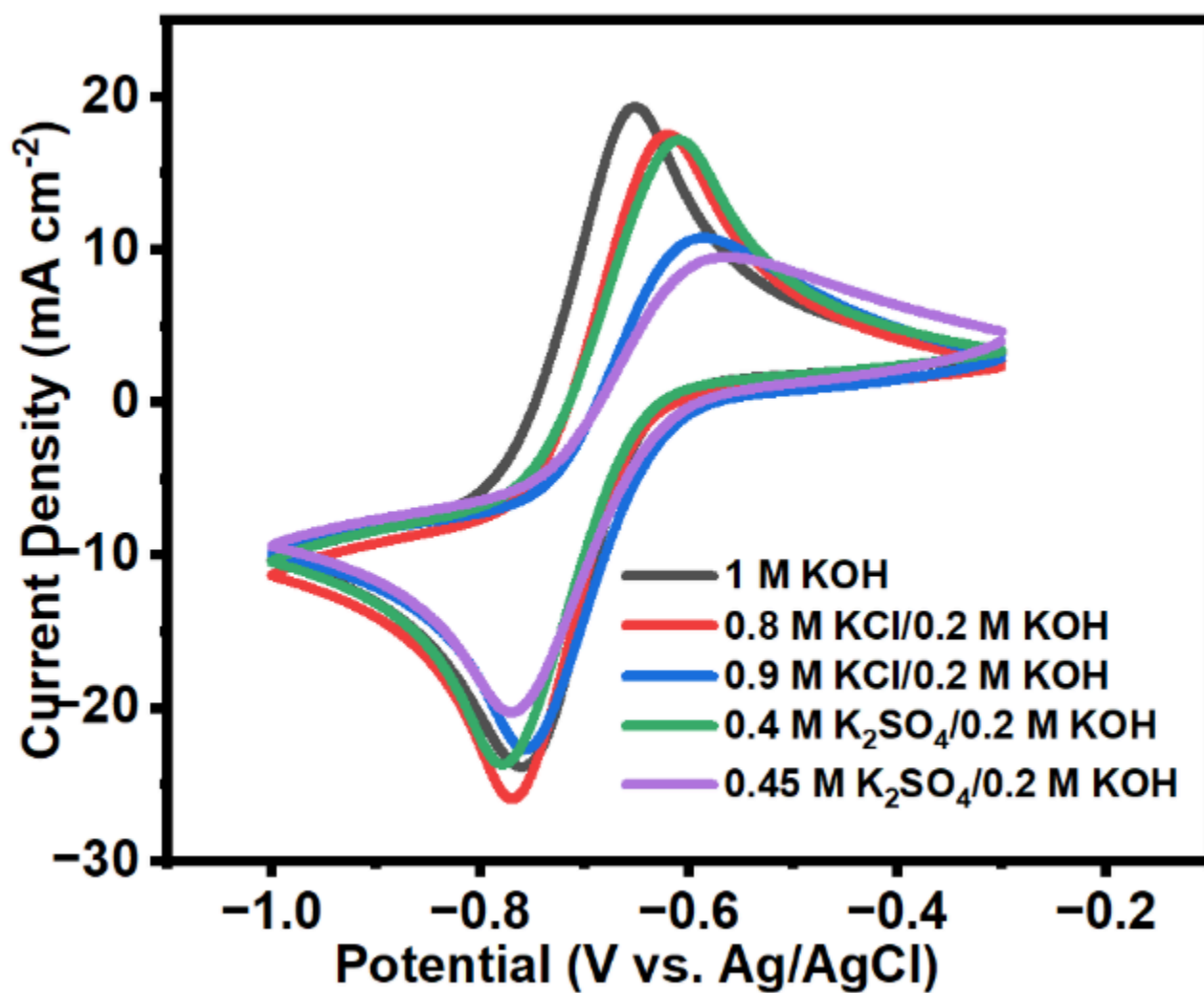


Figure 2

CV curves of 0.1 M Lawsone in i) 1 M KOH, ii) 0.8 M KCl containing 0.2 M KOH, iii) 0.9 M KCl containing 0.1 M KOH, iv) 0.4 M K₂SO₄ containing 0.2 M KOH, or v) 0.45 M K₂SO₄ containing 0.1 M KOH.

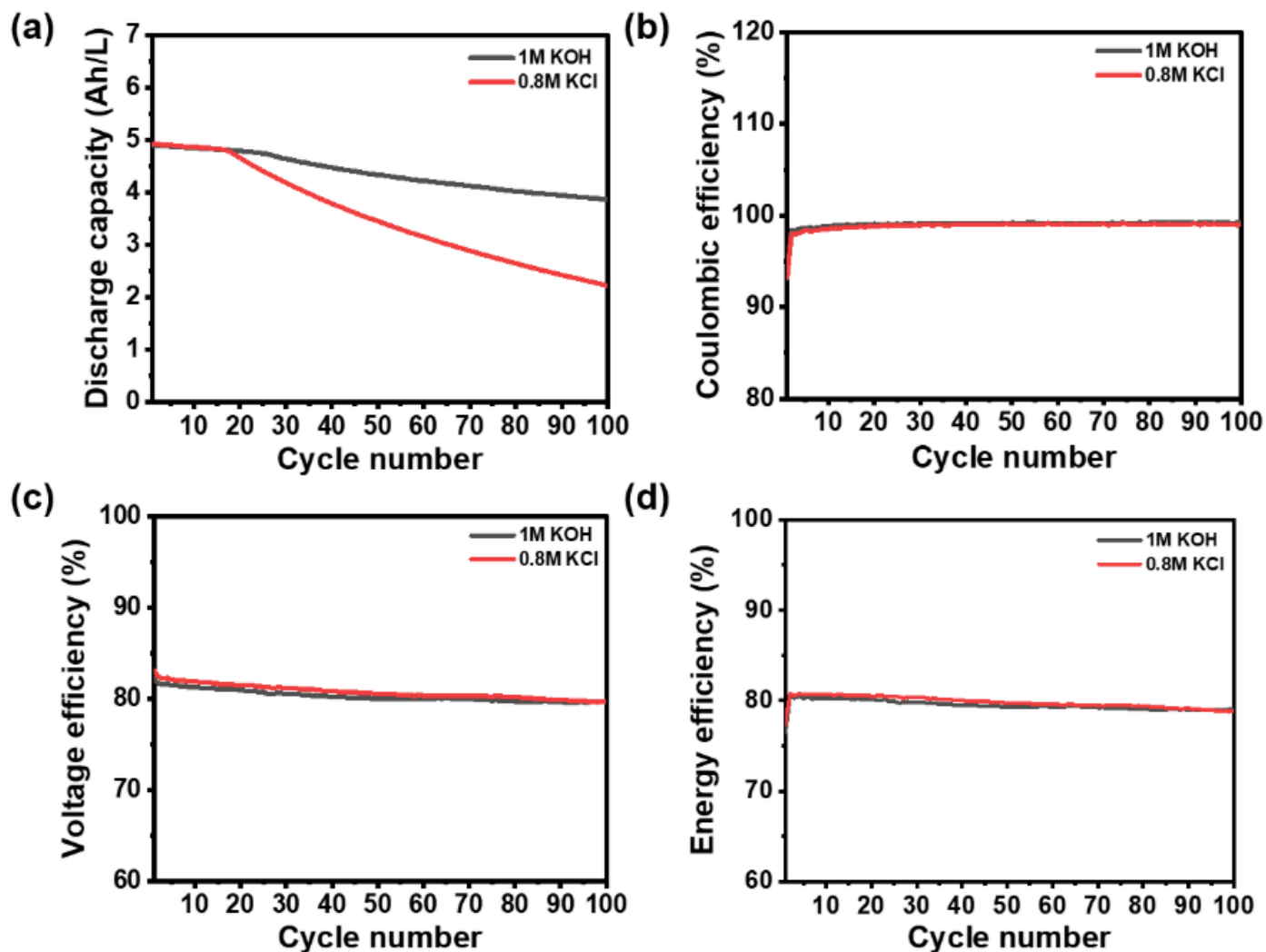


Figure 3

(a) Discharge capacity, (b) Coulombic efficiency, (c) Voltage efficiency, and (d) Energy efficiency of single cells measured with 0.1 M Lawsone in i) 1 M KOH and ii) 0.8 M KCl containing 0.2 M KOH. The tests were performed at 40 mA cm⁻² for 100 cycles.

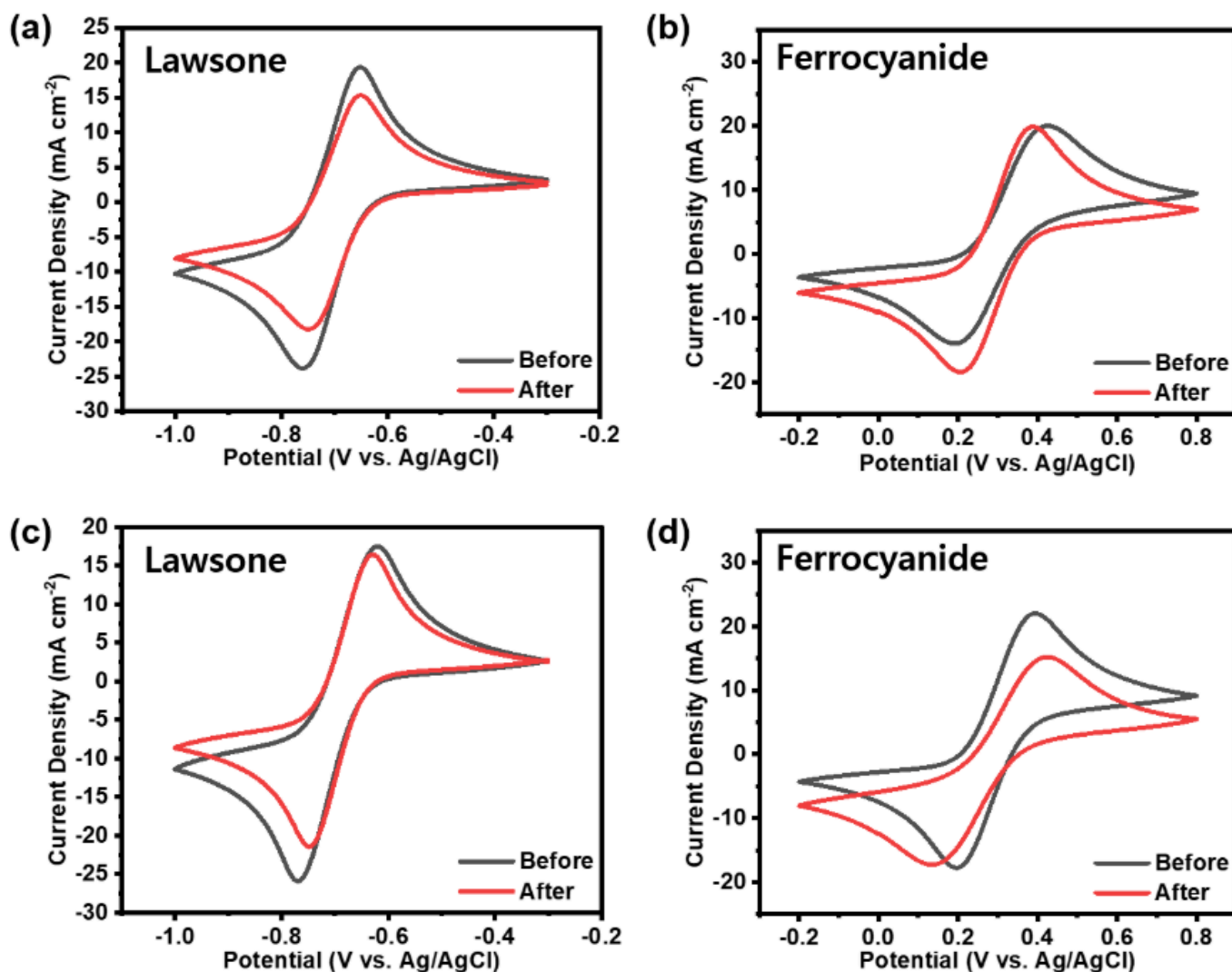


Figure 4

CV curves of (a) anolyte and (b) catholyte before and after single cell test using an anolyte with 1 M KOH, and the (c) anolyte and (d) catholyte before and after single cell test using an anolyte with 0.8 M KCl containing 0.2 M KOH.

Supplementary Files

This is a list of supplementary files associated with this preprint. Click to download.

- [Scheme1.jpg](#)