An aqueous redox flow battery using CO_2 as an active material

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2026.6.17 IFBF2026@Budapest

Research Background

Expanded Deployment of Variable Renewables

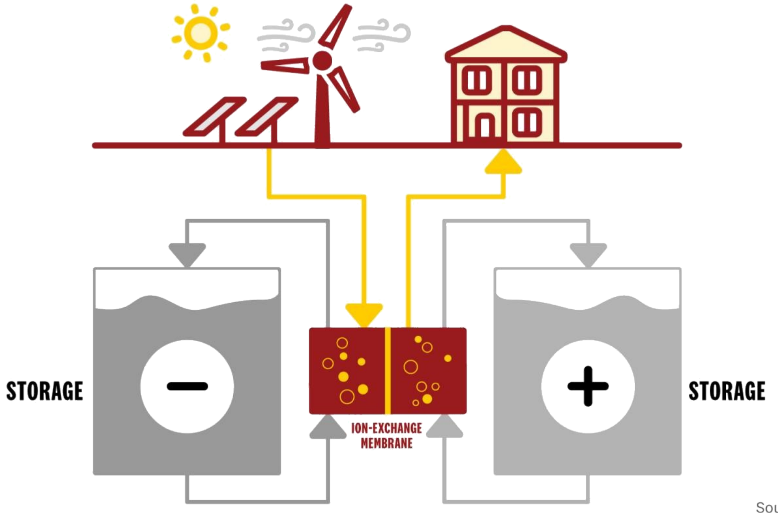
- Grid stabilization is essential as renewables become the main power source

Improving Resilience

- A power source capable of handling long outages from natural disasters is essential

Challenges of Existing Energy Storage Technologies

[Vanadium Redox Flow Battery]



- Inherently safe, with cost advantages estimated for long-duration applications
- **Issue: Resource constraints and geopolitical risks of vanadium**

Toward Next-Generation Redox Flow Batteries

Inorganic active materials

Inexpensive and abundant metals…Mn and Fe etc.

Organic active materials

C ・ H ・ N ・ O as main components (ubiquitous elements)

…Potential, solubility, and stability can be systematically

optimized

via molecular design

Toward Next-Generation Redox Flow Batteries

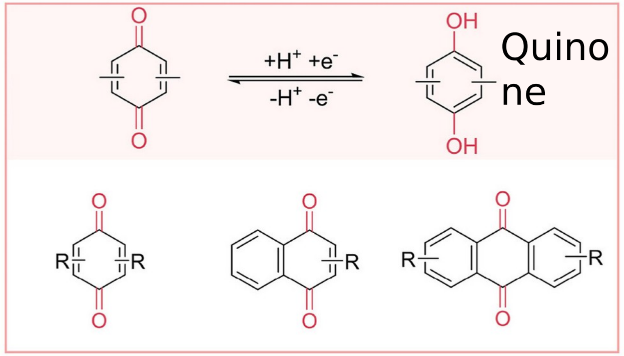
Examples of aqueous organic active materials

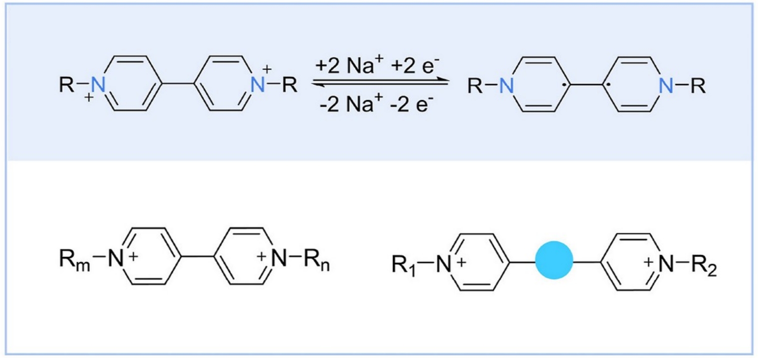
Viologen

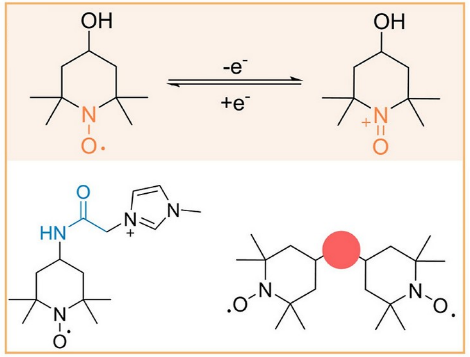
Quinone

TEMPO

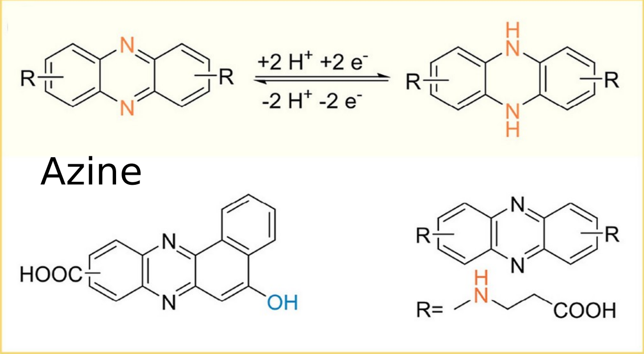
Quino
ne



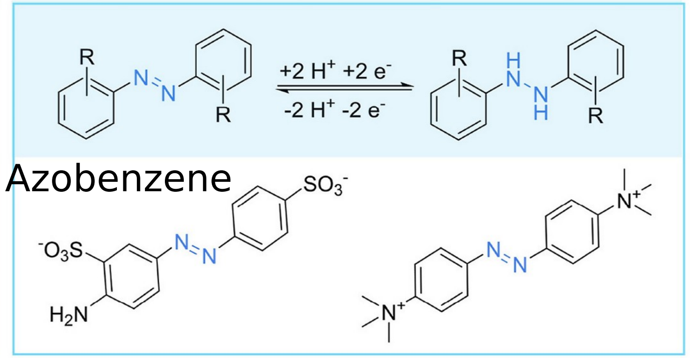




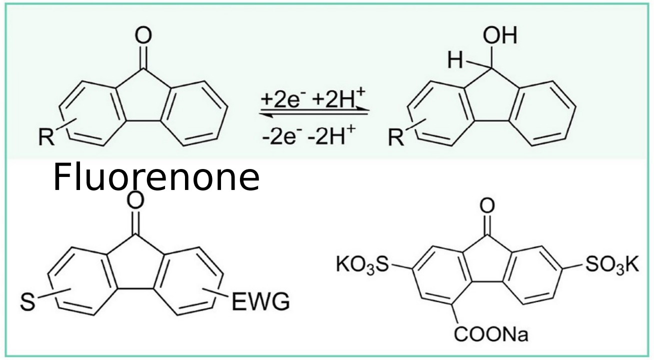
Azine



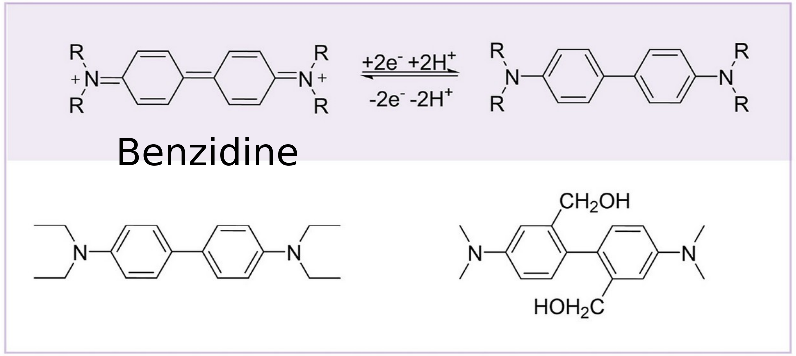
Azobenzene



Fluorenone



Benzidine



F. Zhu et al. *Green Energy Environ.* 9, 1641–1649 (2024).

Toward Next-Generation Redox Flow Batteries

- Limited frameworks for candidate active materials
- Expanding active-material frameworks via catalytic transformations

$$\text{R-OH} \overset{\text{cat.}}{\leftrightarrows} \text{R=O} + \text{H}^+ + \text{e}^-$$

$$\text{R-H} + \text{R'-H} \overset{\text{cat.}}{\leftrightarrows} \text{R-R'} + 2\text{H}^+ + 2\text{e}^-$$

$$\text{Ar-H} + \text{Ar'-H} \overset{\text{cat.}}{\leftrightarrows} \text{Ar-Ar'} + 2\text{H}^+ + 2\text{e}^-$$

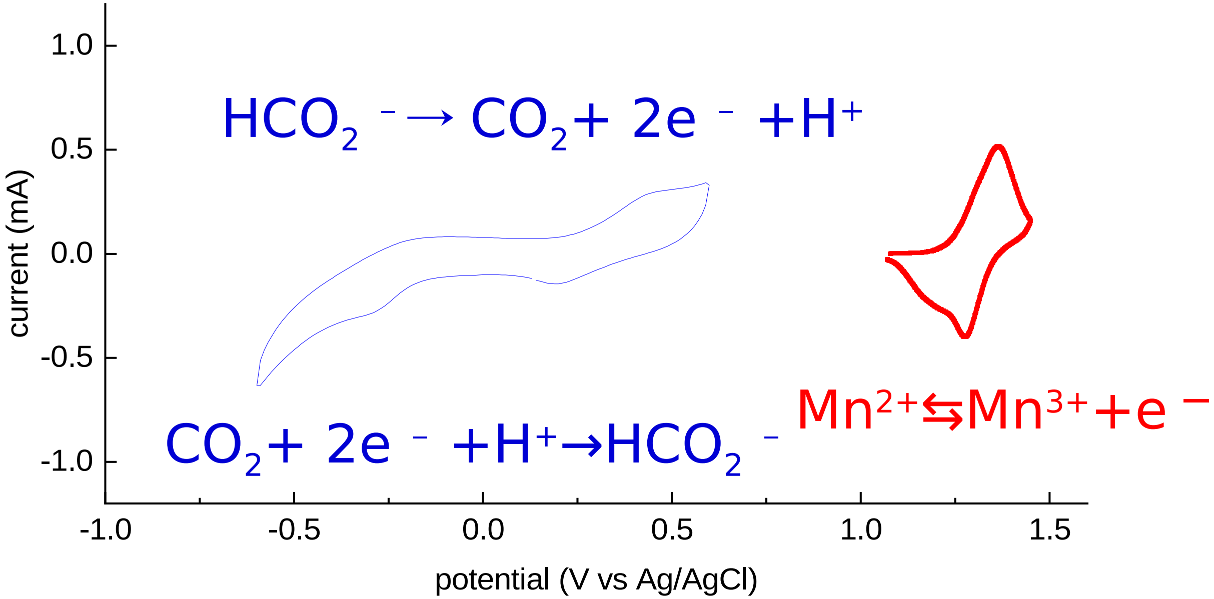
$$\boxed{\text{HCO}_2^- \overset{\text{cat.}}{\leftrightarrows} \text{CO}_2 + \text{H}^+ + 2\text{e}^-}$$

Strategy for constructing battery

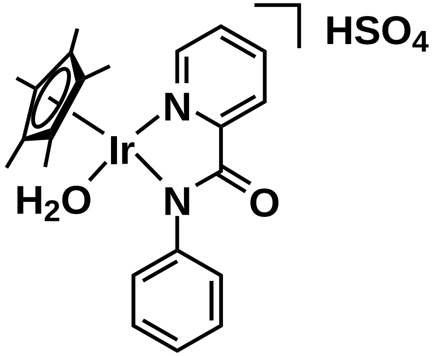
CO_2 electroreduction

$$CO_2 + H^+ + 2e^- \xrightarrow{\text{Catalyst}} HCO_2^-$$

We focused on the Ir catalyst we have developed, which functions in CO_2 electrochemical reduction.



Catalyst



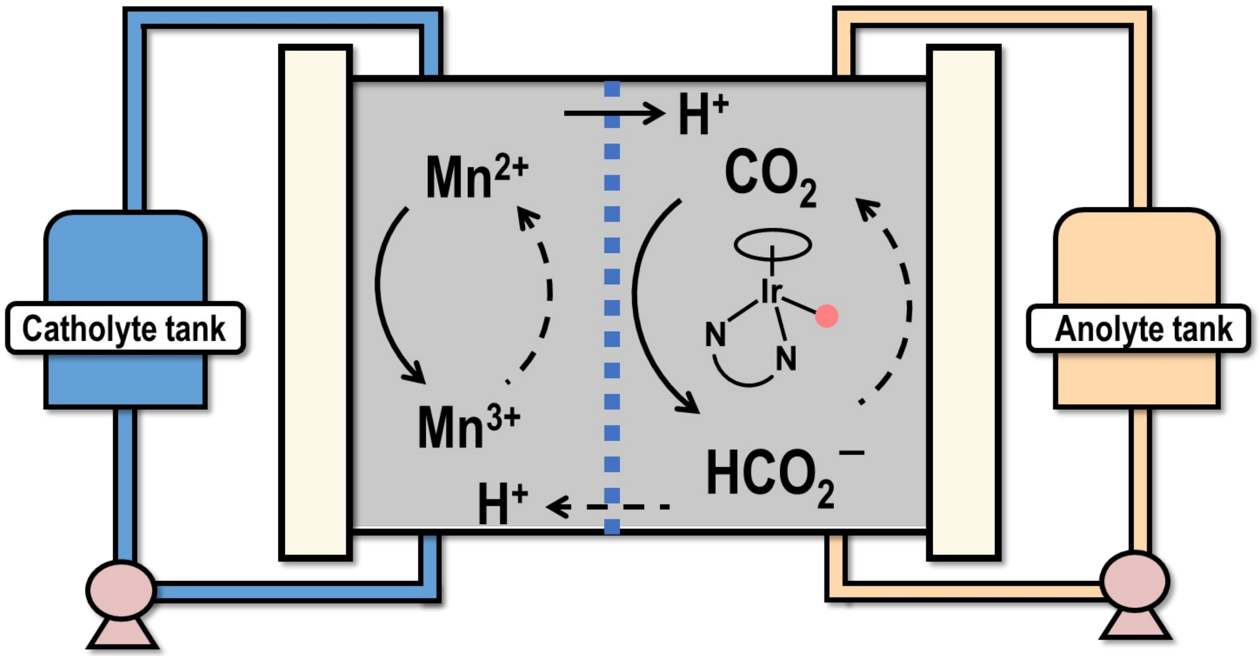
R. Kanega, et al. *ACS Catal.*, **2018**, 8, 11296

2 M $KHCO_3$/Ir complex under CO_2

WE: carbon felt

RE: Ag-AgCl

Experimental



Anolyte $$CO_2 + H^+ + 2e^- \underset{\text{Discharging}}{\overset{\text{Charging}}{\rightleftharpoons}} HCO_2^-$$

Catholyte $$2Mn^{2+} \underset{\text{Discharging}}{\overset{\text{Charging}}{\rightleftharpoons}} 2Mn^{3+} + 2e^-$$

Negative electrolyte

1 mM Ir catalyst/2 M $KHCO_3$

CO_2 (3 mL min^{-1})

Positive electrolyte

20 mM $MnSO_4$/20 mM V^V/3 M H_2SO_4

0.2 M $MnSO_4$/0.2 M V^V/3 M H_2SO_4

Electrode

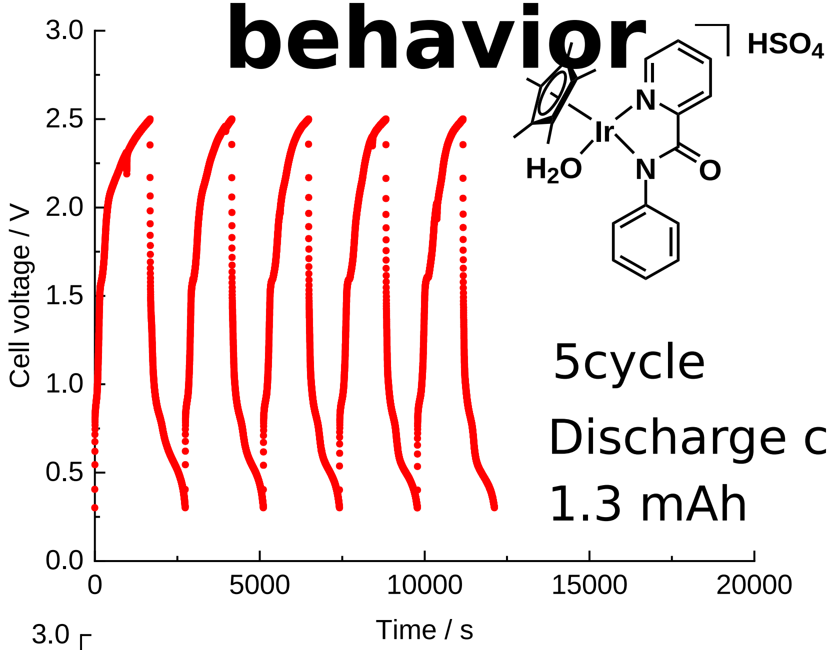
Carbon felt

(1×2.5 cm, AAF304ZS, Toyobo)

Membrane

N117/NR212/N117

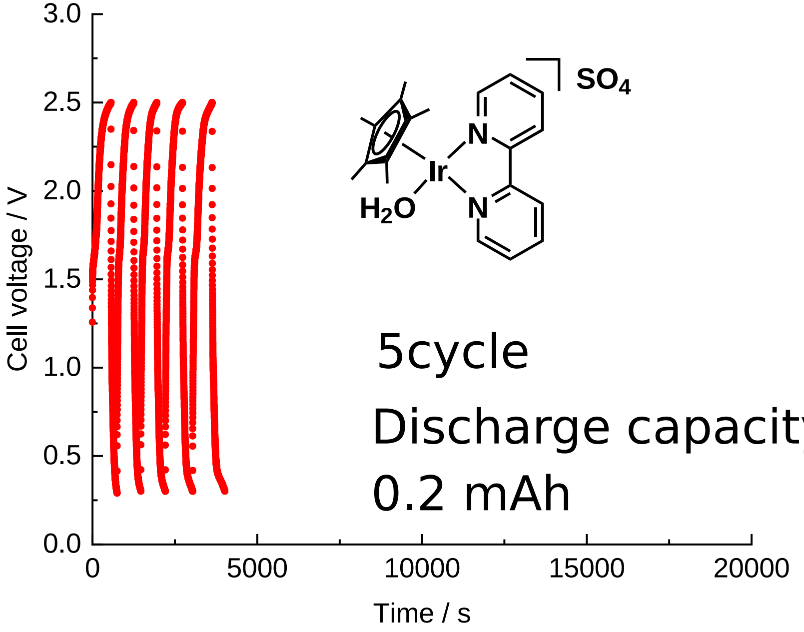
Charge-discharge behavior



5cycle

Discharge capacity

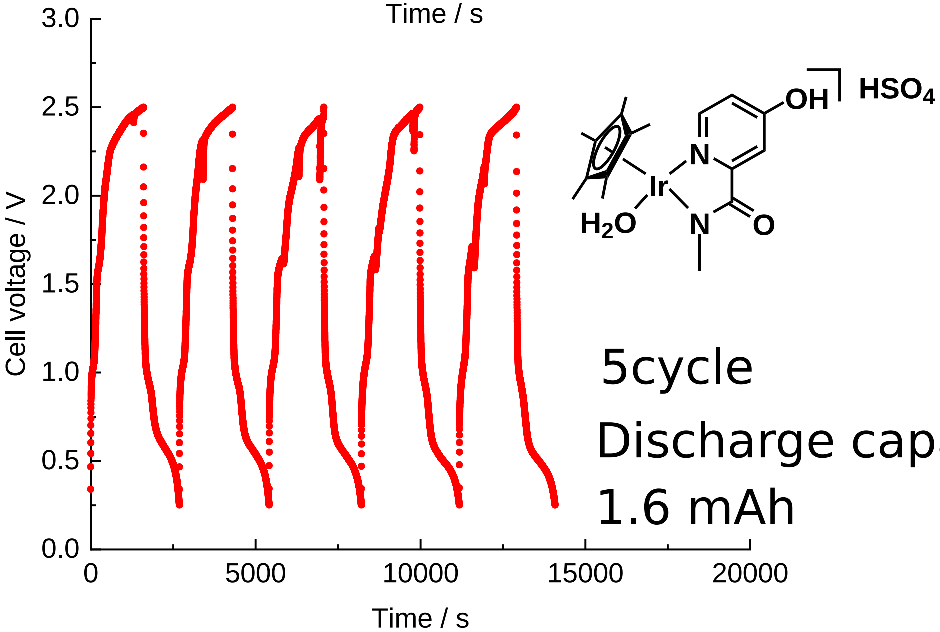
1.3 mAh



5cycle

Discharge capacity

0.2 mAh



5cycle

Discharge capacity

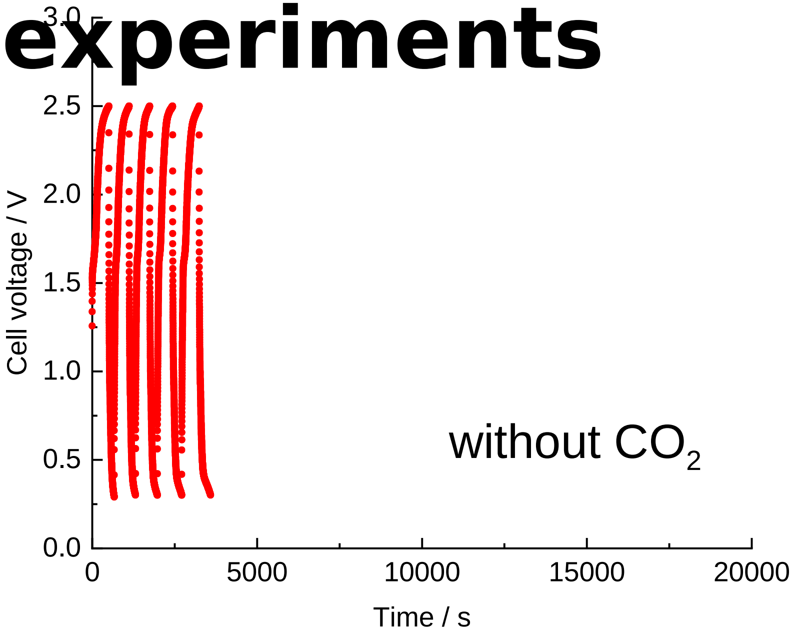
1.6 mAh

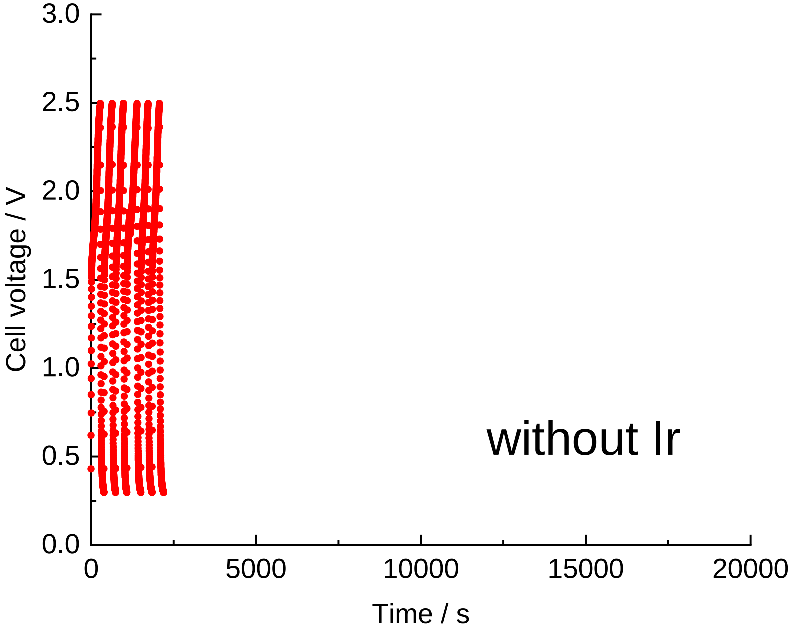
1 mM Ir complex/2 M $KHCO_3$, CO_2 3mL min^{-1}

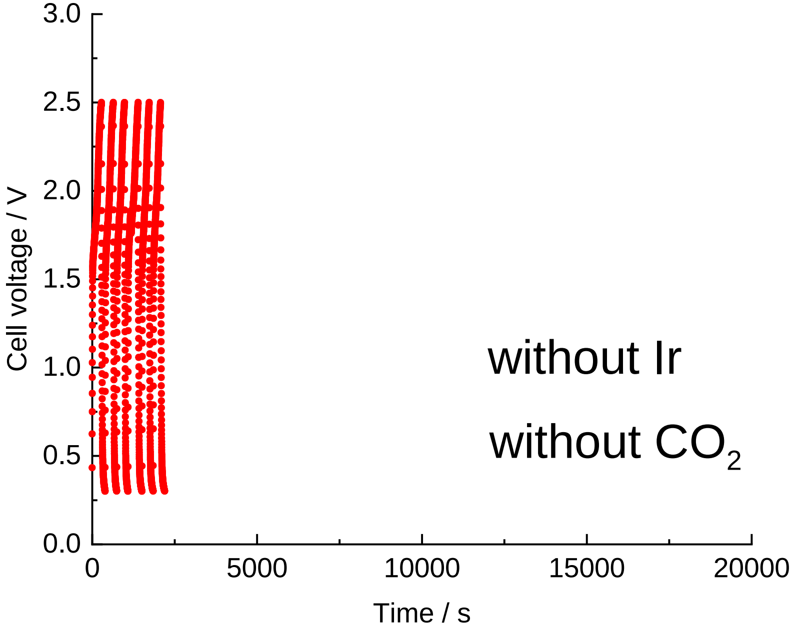
10 mM Mn^{2+}/10 mM V^{5+}/3 M H_2SO_4

Charge and discharge 5 mA

Control experiments

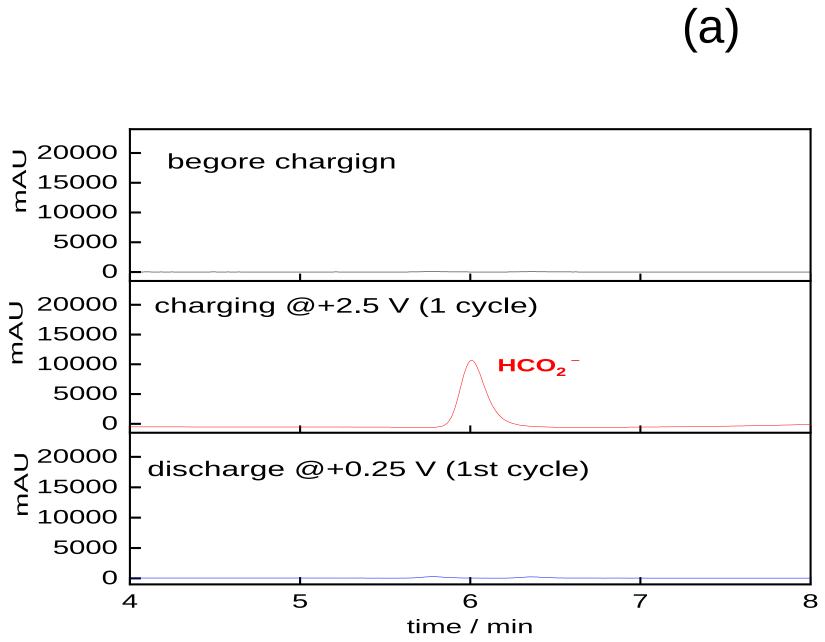


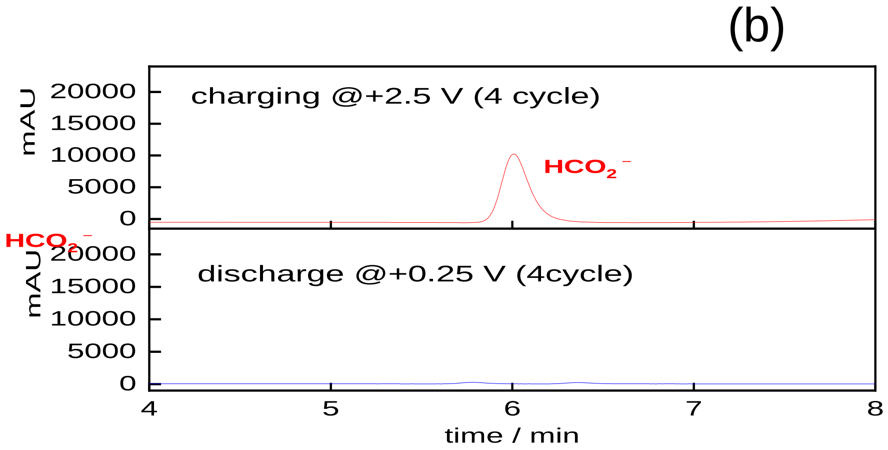


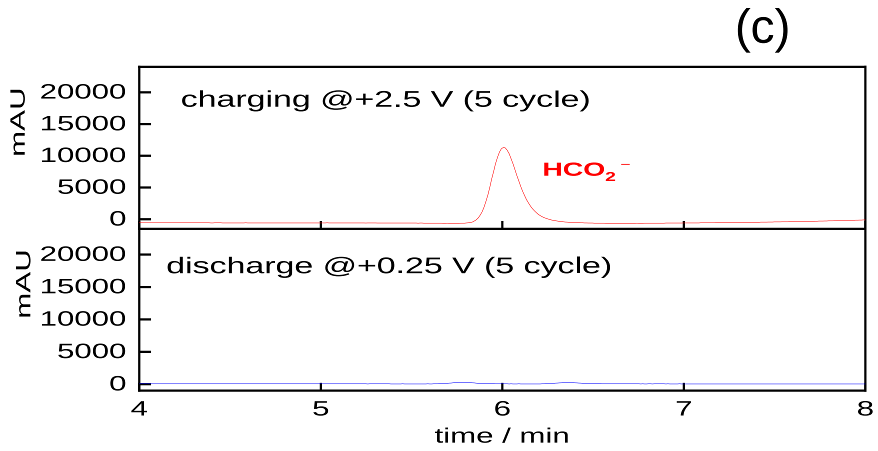


The battery did not work without the Ir catalyst or CO_2.

Negative electrolyte analysis



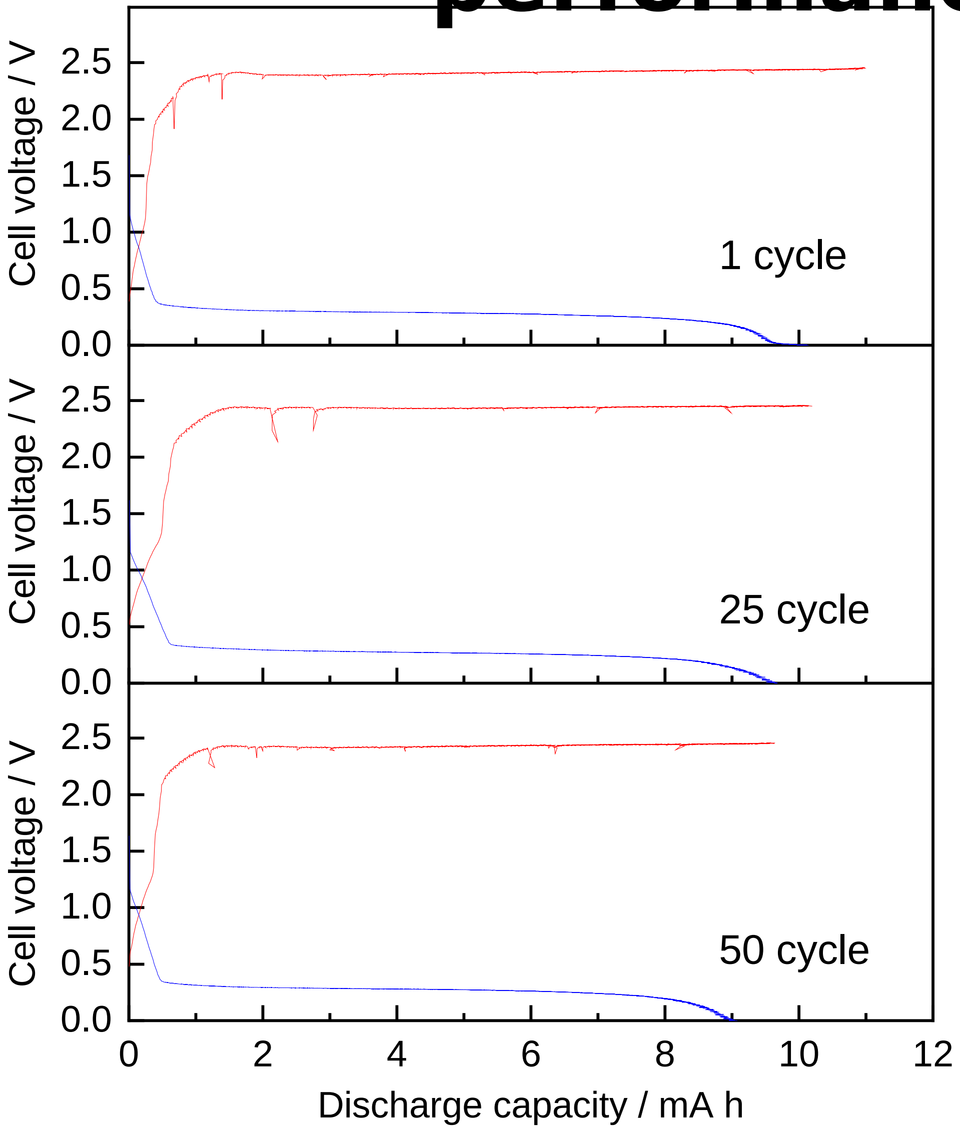


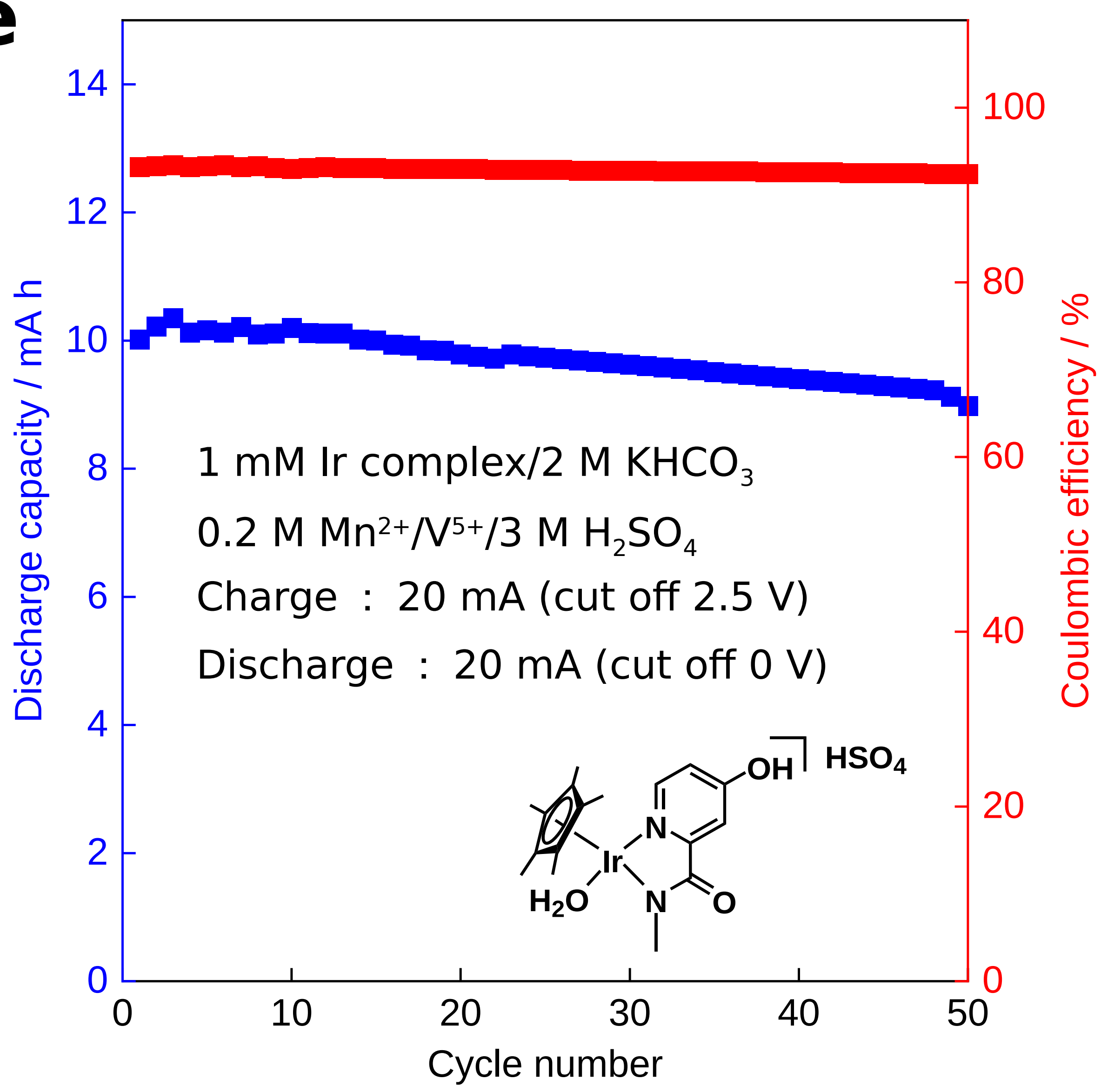


Charge: HCO_2^- generation

Discharge: HCO_2^- consumption

Long-term cycling performance





Summary

We constructed an aqueous RFB using CO_2 as the active material with a homogeneous catalyst. The system gave a maximum discharge capacity of 10.5 mAh (1.5 AhL^{-1}) and capacity decay of 0.2% per cycle. This approach is expected to be adaptable to various active materials via catalytic transformation.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Redox Flow Batteries

How to cite: *Angew. Chem. Int. Ed.* **2023**, *62*, e202310976
doi.org/10.1002/anie.202310976

An Aqueous Redox Flow Battery Using CO_2 as an Active Material with a Homogeneous Ir Catalyst**

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Angewandte Chemie

R. Kanega, et al., *Angew. Chem. Int. Ed.* **2023**, 62, e202384761.

This research is supported by the New Energy and Industrial Technology Development Organization program.