



Automation-Enabled High-Throughput Workflows for Redox Flow Battery Materials Screening and Validation

Leveraging MIRAL Capabilities for DOE OE Redox Flow Battery Screening, Validation, and Down-Selection

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Project Overview

- **Goal:** Develop automation-enabled, high-throughput experimental workflows to accelerate RFB materials screening, formulation optimization, electrochemical validation, and down-selection of lower-cost chemistries.
- **Current Practice:** Discovery still relies heavily on sequential manual experiments, limited-throughput characterization, and slow iteration from materials/formulation screening to cell validation.
- **Why PNNL:** PNNL combines deep redox flow battery expertise with MIRAL's programmable automation, high-throughput experimentation, multimodal characterization, electrochemistry, and structured data capture.
- **Innovation:** MIRAL connects robotic sample preparation, HTE solubility/electrochemical screening, targeted cell testing, and decision-quality data into a reusable screening-to-validation workflow.
- **Duration:** FY25–FY26; focus is on reusable workflows, validated datasets, and practical tools that support current and future DOE OE redox flow battery projects.
- **Impact:** Enables faster identification of promising lower-cost chemistries, reduces experimental bottlenecks, improves data quality, and supports national labs, DOE OE, and flow battery developers.
- **Vertical/Investment Area:** Analytics; Materials and Systems Innovation; Safety & Reliability
- **Alignment/Supports strategic pathway:** Supports DOE OE goals to grow grid capacity, stabilize grid services, and improve reliability through scalable, lower-cost long-duration energy storage technologies.

Motivation: Faster Materials-to-System Translation for Energy Storage



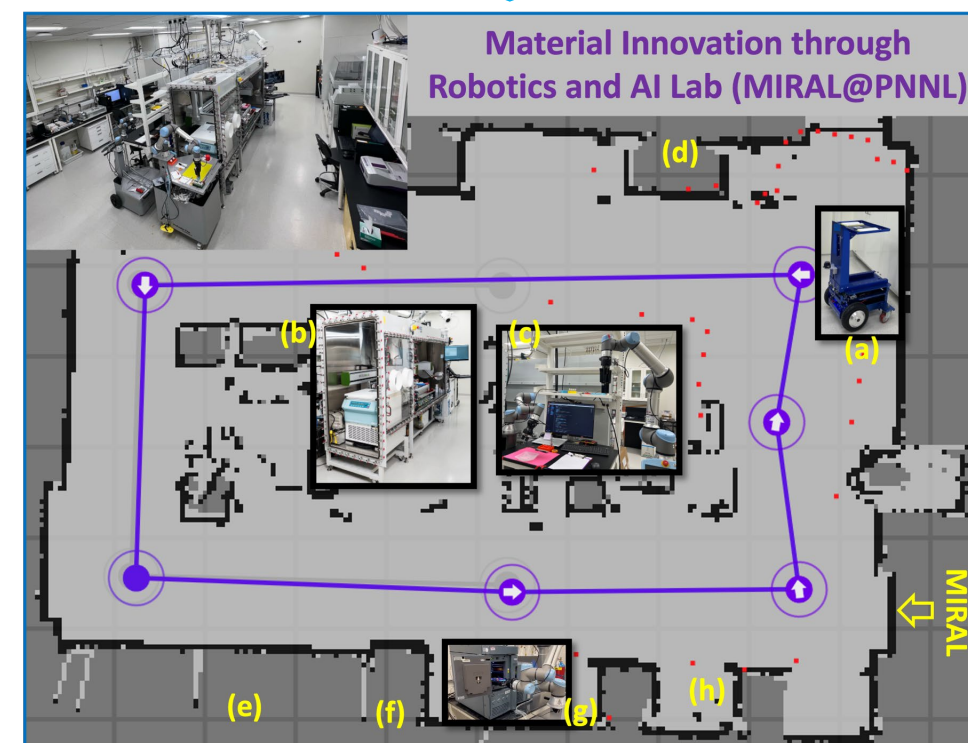
Urgent need: grid-scale energy storage

- Rising electricity demand from data centers, electrification, and industrial growth
- Storage improves grid reliability, resilience, security, and affordability
- Redox flow batteries are promising for long-duration storage

Faster materials screening and validation are needed to deliver grid-ready storage solutions.

Bottleneck: slow materials-to-system translation

- **Large materials/formulation spaces:** redox-active molecules, salts, solvents
- **Coupled property requirements:** solubility, conductivity, stability
- **Fragmented characterization and validation:** spectroscopy, CV/EIS, cycling
- **Sparse trusted datasets:** limited standardized data and metadata
- **Slow scale-up:** formulation → cell → stack



MIRAL accelerates screening, validation, and down-selection through automation-enabled HTE workflows.

High-Throughput Capabilities at PNNL — MIRAL

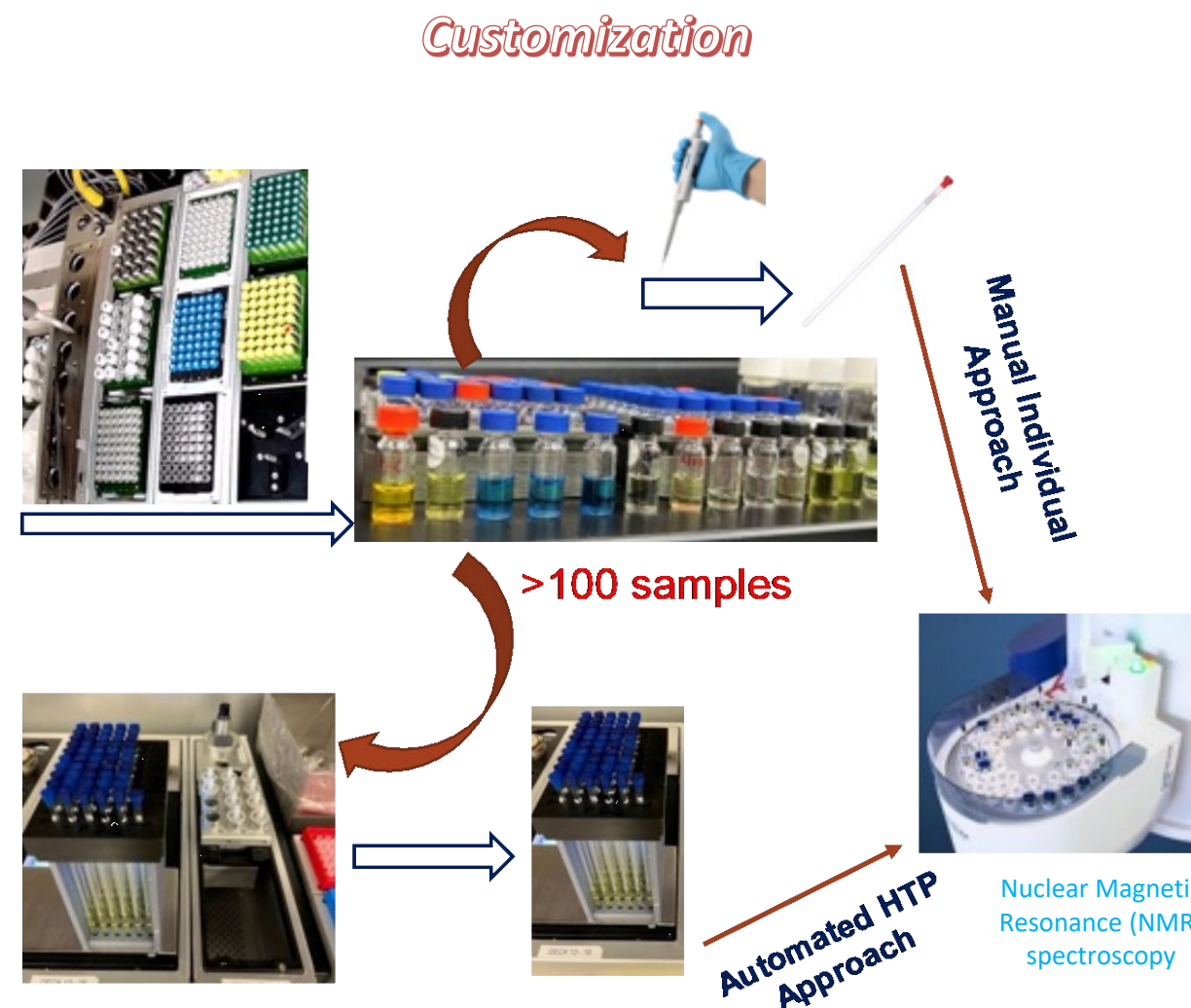
HTP Synthesis & Sampling



Under an inert atmosphere of nitrogen (Archer) or argon (Beverly)

- Multi-functional hardware
- Workflow control software
- Easy integration with end-user workstations

HTP/Automated Characterization



Customizing instruments or workflow to align with project requirements

MIRAL Workflow Engine: From Integrated Hardware to Reusable HTE Workflows



- 1 HTP solubility workflow
- 2 HTP viscosity and automated imaging
- 3 HTP electrochemical characterization
- 4 Fluorenone solubility screening
- 5 Carbazole catholyte additive screening

Integrated automation, characterization, data capture, and domain expertise enable reusable workflows for energy-storage materials screening and validation.

HTP Workflow for Thermodynamic Solubility Determination



Robotically controlled platform with a HTP methodology



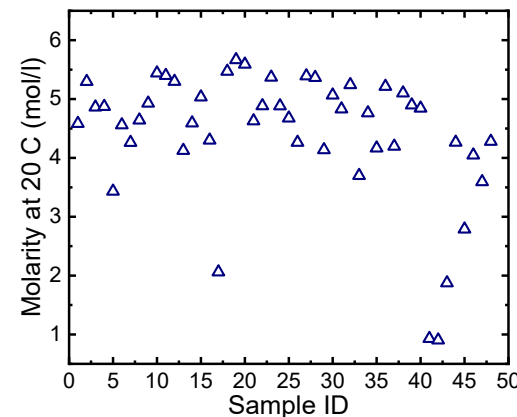
Step 1:
*Mixing and
Equilibration*

Step 2:
*Filtration or
Centrifugation*

Step 3:
*Sample
Preparation*

Step 4:
*Analysis (NMR
or UV/VIS)*

Follow the classical shake-flask method



Quantity



Quality

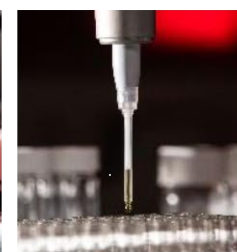
- Sufficient equilibration time is needed to reach true saturated solutions
- Running numerous parallel experiments to increase throughput (>200 samples/round experiment)
- Automation improves throughput, consistency, and expert-driven validation

HTP Viscosity and Automated Imaging

HTP Physical-Property Screening: Viscosity, Imaging, and Stability Indicators

HTP Viscosity Screening

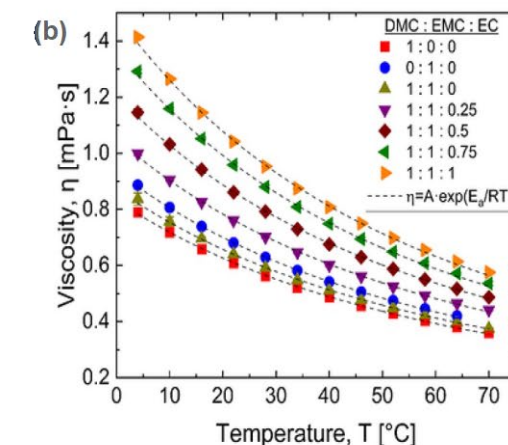
- ~100 μL /sample, 40-vial plate format
- Rapid viscosity–temperature mapping
- Identifies practical formulation windows
- Guides electrochemical down-selection



40 vials/plate

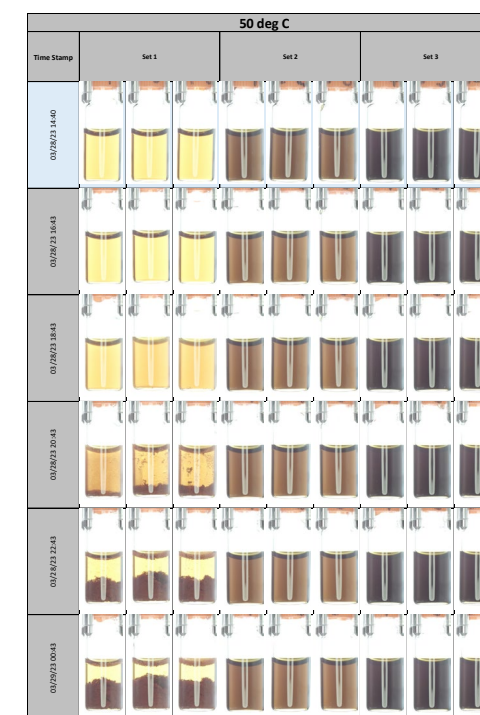
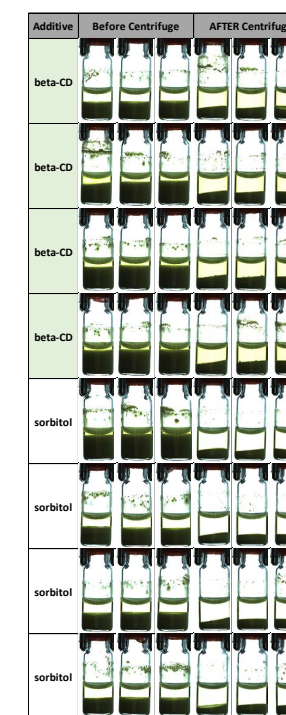


~100 μL /sample



Automated Imaging Capability

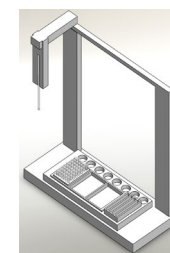
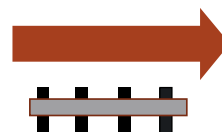
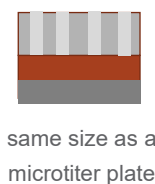
- Integrated robotic imaging and sample handling
- Tracks precipitation, phase separation, color, and aging
- Converts sample images into analyzable datasets
- Supports automated analysis and formulation decisions



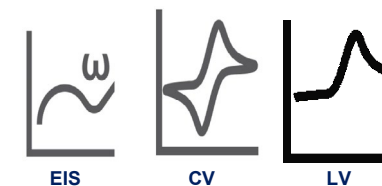
HTP Electrochemical Characterization



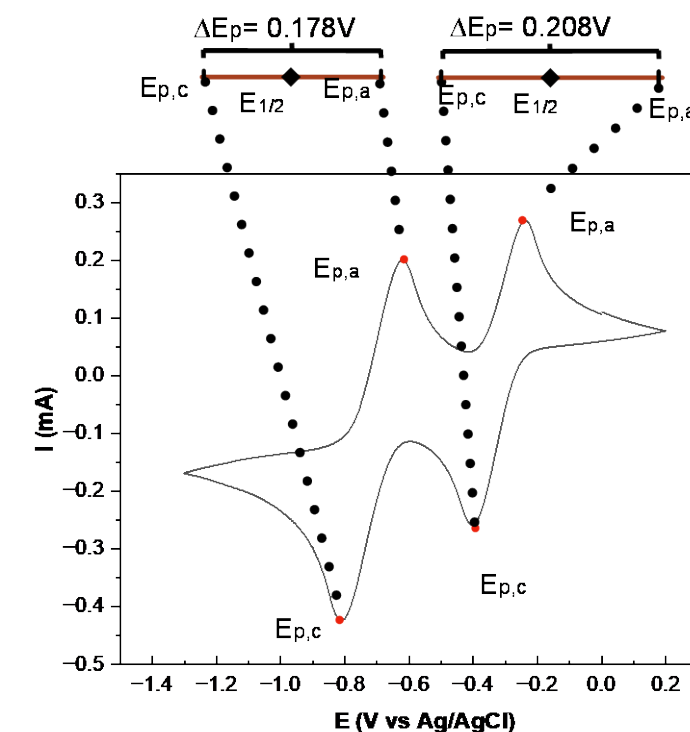
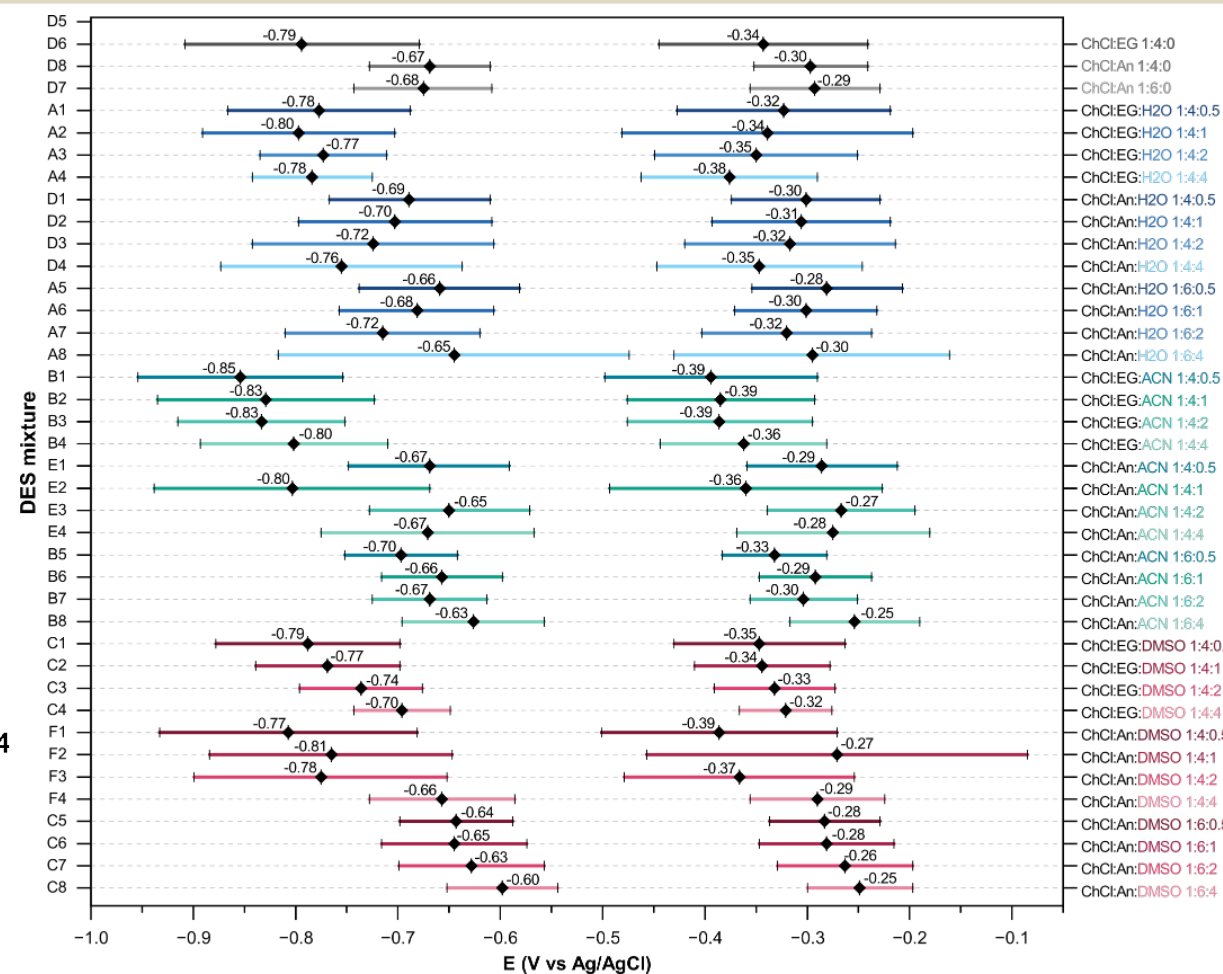
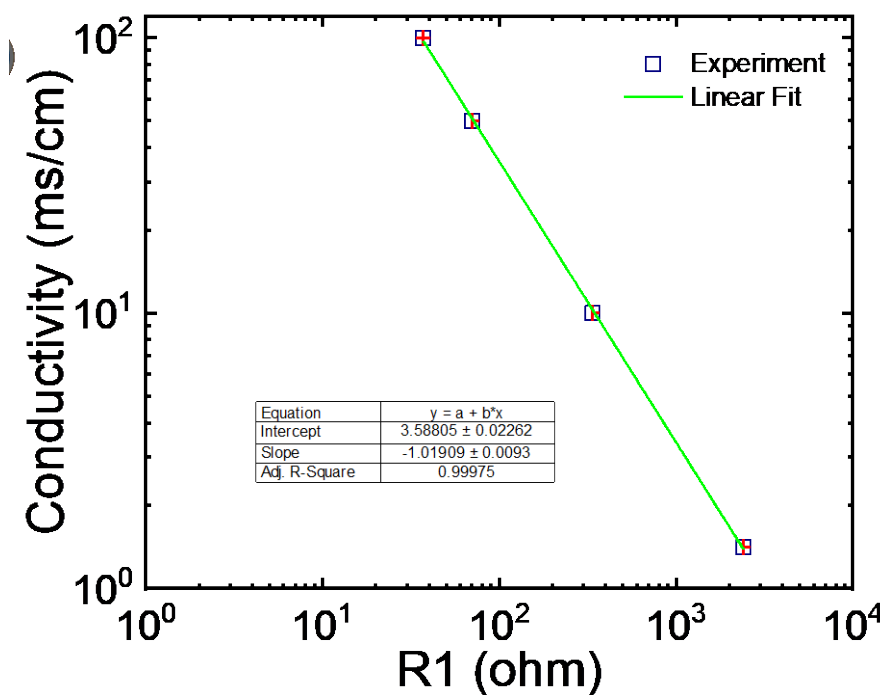
1. Automatic sampling



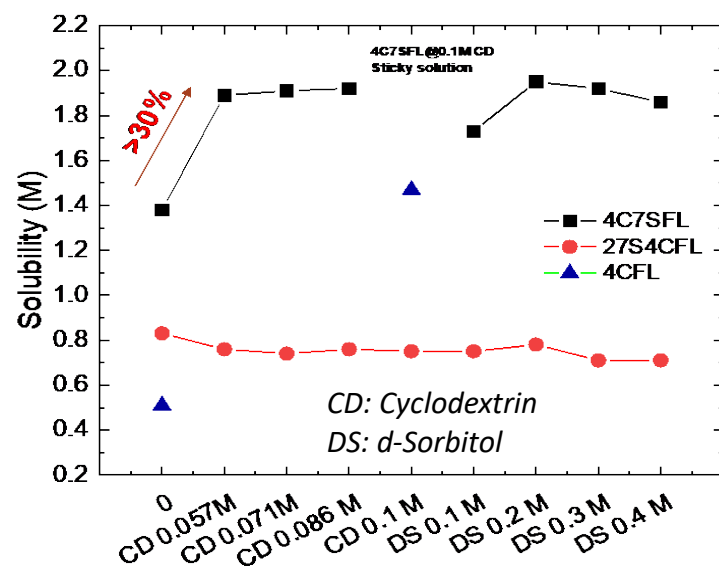
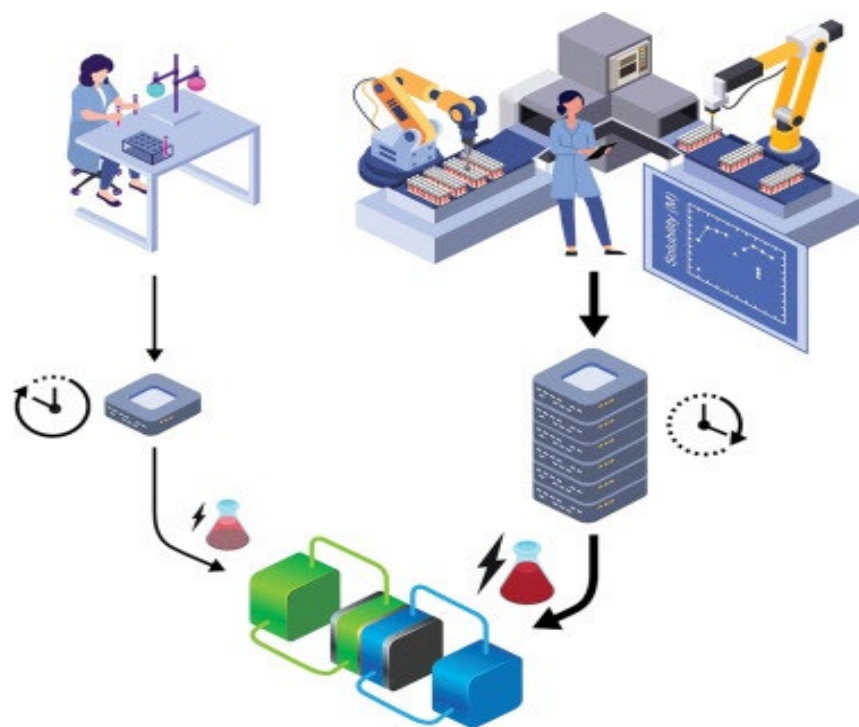
2. Measurement setup



3. Data collection and analysis

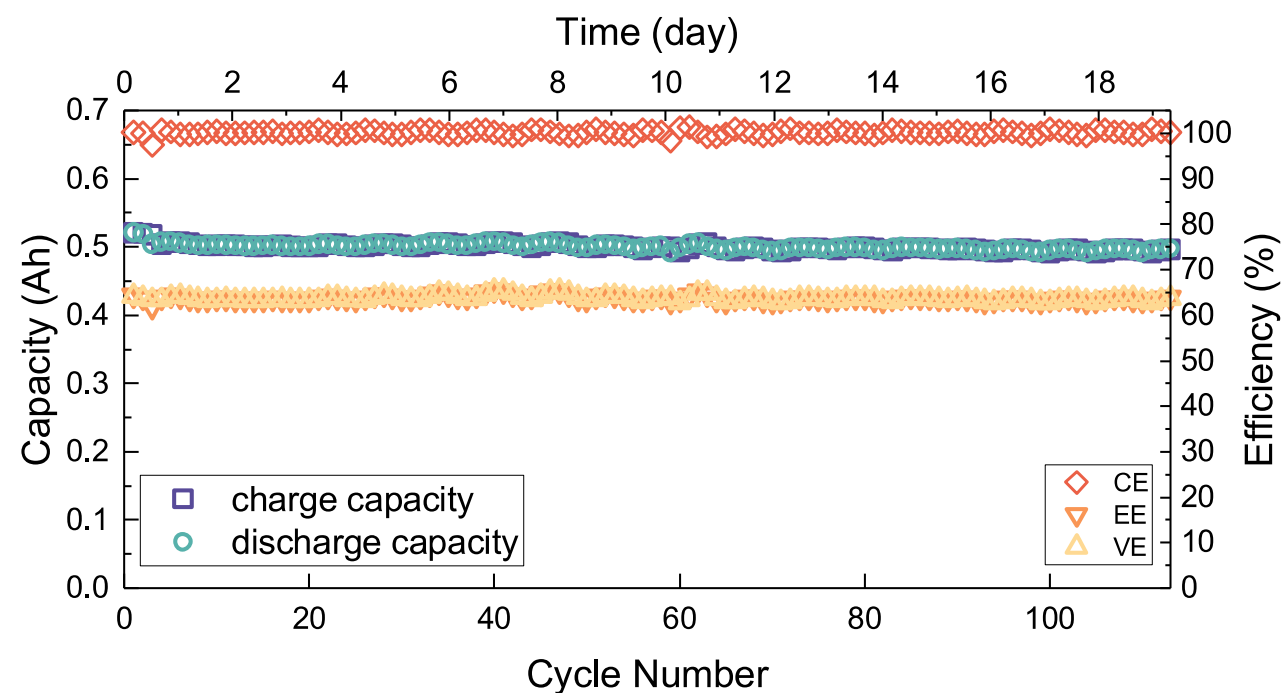


High-quality Solubility Data for Aqueous Redox Flow Battery Research



Highlights

- Automated high-throughput solubility determination of redox-active materials
- Standardized high-throughput workflow for large-scale, high-quality solubility data
- Studied *the effects of additives* on solubility in aqueous redox flow batteries
- Highly stable performance over 18 days with consistent capacity and a 24% boost in energy density

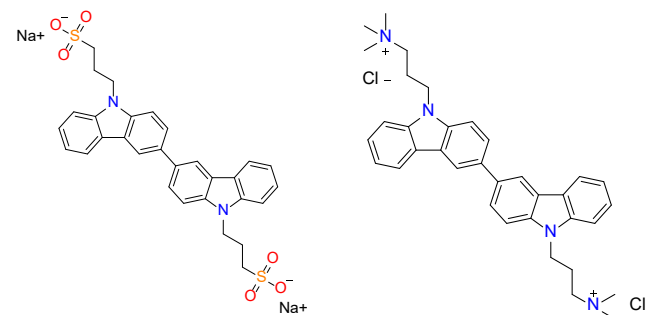


MIRAL Accelerates Testable Catholyte Formulations, but Cell-Level Bottlenecks Remain

From formulation improvement to cell-level translation challenges

Low-solubility bi-Cz catholyte → testable formulations

- Expert-selected low-solubility bi-Cz catholyte
- MIRAL HTE additive screening and image-based ranking
- NA/TMBAC leads: ~3× solubility increase
- No obvious barriers from CV, conductivity, or viscosity



Flow-cell testing reveals translation limits

- Degradation
- Crossover
- pH drift
- Poor discharge reversibility

Code	Chemical / formulation aid	Primary function	Key outcome
SAA	Sulfanilic acid	Aromatic sulfonate hydrotrope	Poor
NaTS	Sodium p-toluenesulfonate	Aromatic sulfonate hydrotrope	Poor
Gly	Glycine	Zwitterion / amino acid	Poor
β-Ala	β-Alanine	Zwitterion / amino acid	Poor
Arg-Cl	DL-Arginine chloride	Cationic amino acid	Poor
Urea	Urea	Weak hydrotrope	Weak
NA	Nicotinamide	Hydrotrope; π-compatible solubilizer	3–4× gain; strong for QA
TMBAC	N,N,N-Trimethylbutan-1-ammonium chloride	Cationic supporting electrolyte; ion-pair modifier	>0.1 M C3-SO3Na
NA+TMBAC	Nicotinamide + TMBAC	Combined hydrotrope + cationic matrix	High solubility; C3 discharge poor
Na ₂ SO ₄	Sodium sulfate	Supporting electrolyte	Works with C4 state; stresses C3
NaCl	Sodium chloride	Supporting electrolyte	Salt-out risk
CTAC	Cetyltrimethylammonium chloride	Cationic surfactant / micelle	Positive clue; not preferred
Choline Cl	Choline chloride	Hydrophilic QA salt	Not fully tested alone
Na-HOBS	Sodium 4-hydroxybutane-1-sulfonate	Small sulfonate; state modifier	Mechanistic clue
MeOH/H ₂ O	Methanol/water	Processing / reconstitution solvent	Regenerates C4 gel state
DMSO/H ₂ O	DMSO/water	Strong processing solvent	C3 collapses on water addition



Next need: flexible HTP flow cells and real-time monitoring to diagnose coupled electrolyte–cell effects.

Analysis and Monitoring Technologies for Flow Battery Diagnostics

Real-time electrolyte insight for solubility, composition, and SOC-relevant tracking

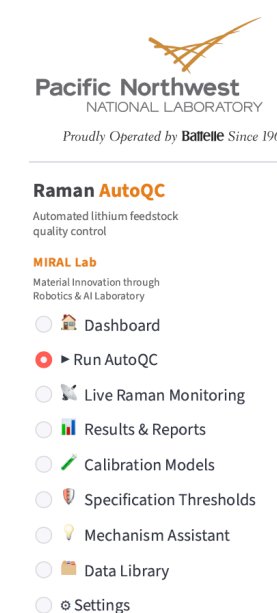
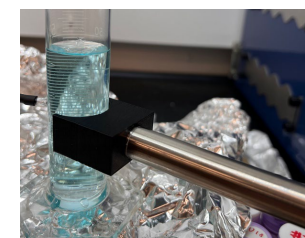
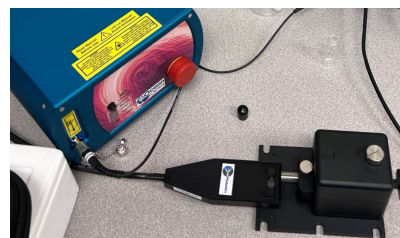
Acoustic diagnostics for Fe–Cr systems

- Non-invasive electrolyte / system monitoring
- Adapted from VFB acoustic sensing work (X. Zang et al, *Small methods*, 2019)
- **Limitation:** weak Fe-side SOC sensitivity

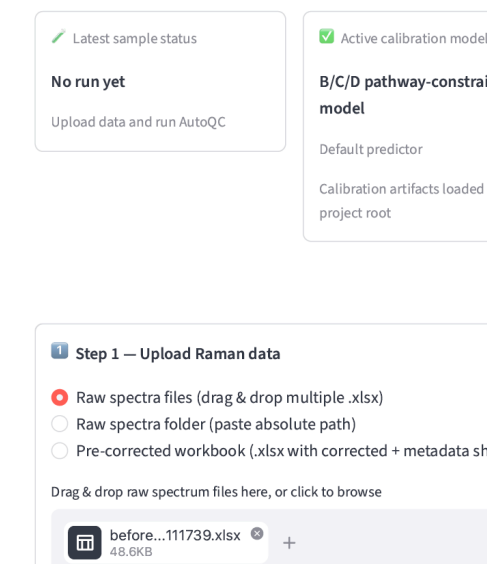
Low attenuation coefficient limits Fe-side SOC diagnosis by acoustic sensing.

Portable Raman + AutoQC for electrolyte diagnostics

- Operando Raman studies show feasibility for tracking RFB electrolyte-state changes
- MIRAL adds portable Raman + automated spectral processing + AutoQC software
- Target outputs: solubility, composition, SOC, and degradation indicators
- Goal: convert spectra into actionable electrolyte diagnostics



Run Raman AutoQC



AutoQC software screenshot

Portable Raman + AutoQC converts electrolyte spectra into solubility, composition, and SOC-relevant diagnostics.

Toward Integrated Workflows for RFB Experimentation

Connecting experiment design, execution, transport, characterization, analysis, and modeling

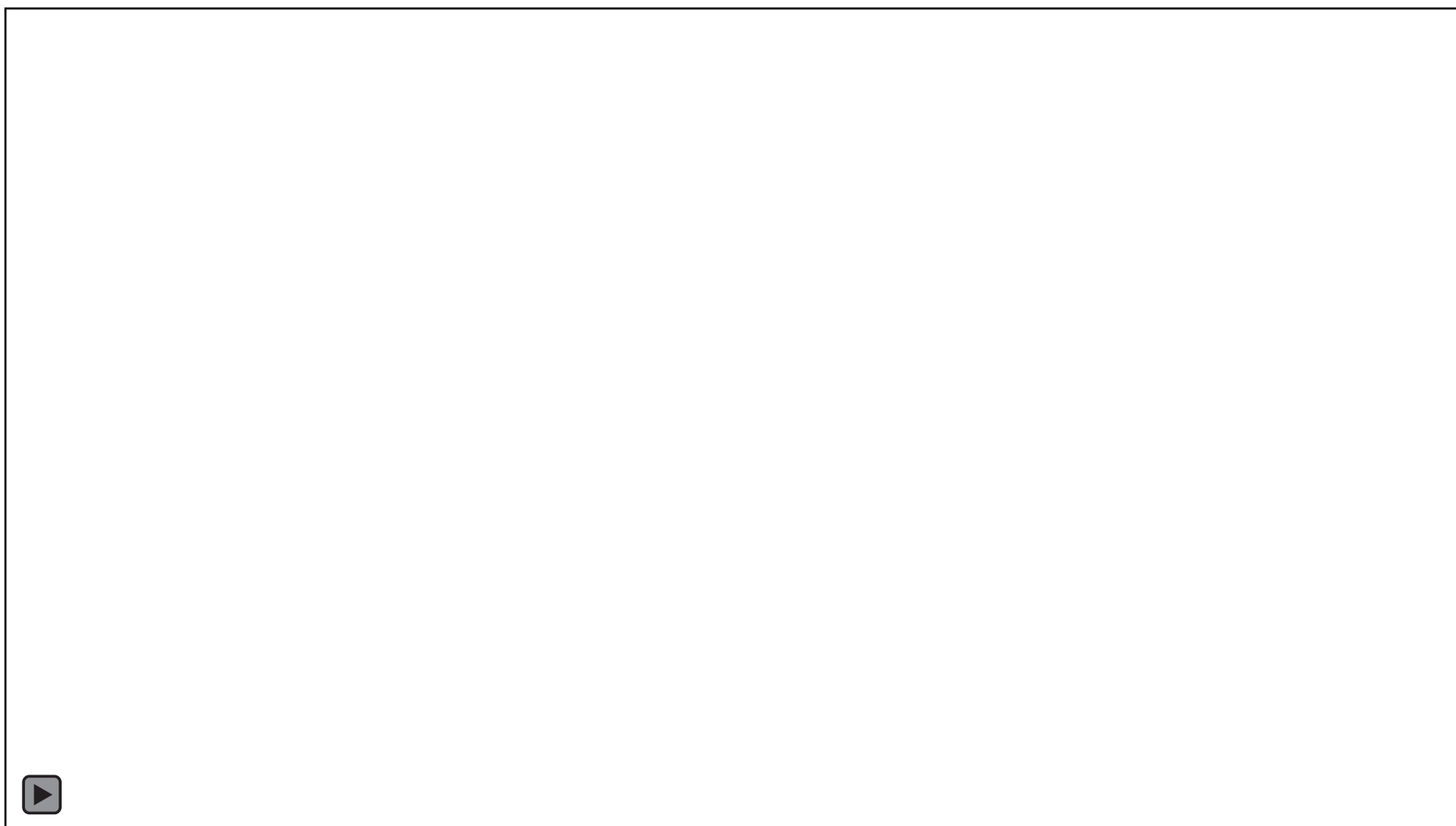
Summary:

Automation-enabled HTE workflows support faster, data-driven RFB materials screening and validation.

Future work:

Integrated MIRAL workflows will connect sample preparation, robotic execution, sample transport, characterization, data analysis, and modeling to support reusable screening-to-validation pipelines.

Demonstration: Design → Execution → Transport → Characterization



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Thank you

