Nonaqueous redox flow batteries (NAqRFBs) are a promising, but nascent, concept for cost-effective grid-scale energy storage. While most studies report new active molecules and proof-of-concept prototypes, few discuss cell design. The direct translation of aqueous RFB design principles to nonaqueous systems is hampered by a lack of materials-specific knowledge, especially concerning the increased viscosities and decreased conductivities associated with nonaqueous electrolytes. To guide NAqRFB reactor design, recent techno-economic analyses have established an area specific resistance (ASR) target of <5 Ω cm². Here, we employ a state-of-the-art vanadium flow cell architecture, modified for compatibility with nonaqueous electrolytes, and a model ferrocene-based redox couple to investigate the feasibility of achieving this target ASR. We identify and minimize sources of resistive loss for various active species concentrations, electrolyte compositions, flow rates, separators, and electrode thicknesses via polarization and impedance spectroscopy, culminating in the demonstration of a cell ASR of ca. 1.7 Ω cm². Further, we validate performance scalability using dynamically similar cells with a ten-fold difference in active areas. This work demonstrates that, with appropriate cell engineering, low resistance nonaqueous reactors can be realized, providing promise for the cost-competitiveness of future NAqRFBs.

Grid-scale energy storage has emerged as a critical technology for alleviating the intermittency of renewables, improving the efficiency of the existing grid infrastructure, and offering frequency or voltage regulation. Redox flow batteries (RFBs) are particularly attractive storage devices for energy-intensive grid applications. In a typical RFB, charge-storing active species are dissolved in liquid electrolytes, which are housed in large and inexpensive tanks. The electrolytes are pumped through a power-generating electrochemical reactor, wherein the active species undergo reduction or oxidation on the surfaces of porous electrodes to charge or discharge the battery. This unique architecture offers several advantages over conventional battery systems, including decoupled power and energy scaling, long operational lifetimes, easy maintenance, simplified manufacturing, and high active-to-inactive materials ratio (particularly at long storage durations). Despite many attractive features, state-of-the-art RFBs are currently too expensive for widespread adoption. While the majority of RFB electrolytes are aqueous, transitioning to nonaqueous chemicals could enable higher cell potentials via wider electrolyte activities. Significant differences in the physicochemical and electrochemical properties of aqueous and nonaqueous electrolytes lead to uncertainty in how to effectively design flow cells for NAqRFBs. In particular, nonaqueous electrolytes can exhibit markedly different conductivities and viscosities, and active species for NAqRFBs typically exhibit faster reaction kinetics. AQFB development has recently benefited from a series of studies implementing a single electrolyte diagnostic flow cell technique to systematically evaluate cell performance. A schematic of this single electrolyte technique is provided in Figure 1a, where a flow cell is connected to a single electrolyte reservoir at 50% state-of-charge (SOC). The electrolyte passes through the positive side of the cell, where the active species are oxidized, and then loops back through the negative side of the cell, where the charged species are reduced. Since the same active species are present on both sides of the cell, crossover will not adversely impact cell lifetime or convolute data analysis. Additionally, the reservoir SOC is constant and effectively permits cell-level polarization and electrochemical impedance spectroscopy measurements at a steady-state. This technique most accurately mimics a RFB at 50%SOC when the conversion (f) through one side of the single electrolyte cell is low. The single pass conversion is described in Equation 1, where I is the total current (A), $C_{red}$ is the concentration of reduced species in the reservoir (mol m⁻³), n is the number of electrons transferred (-), F is the Faraday constant (C mol⁻¹), and Q is the electrolyte volumetric flow rate (m³ s⁻¹). Figure 1b illustrates how the electrolyte SOC will vary inside the flow cell as a function of normalized path length for various conversions.

$$ f = \frac{I}{C_{red}nFQ} $$

Single electrolyte studies rely on a stable active material; for example, prior AqRFB studies with this technique have exploited the

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**Towards Low Resistance Nonaqueous Redox Flow Batteries**

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Nonaqueous redox flow batteries (NAqRFBs) are a promising, but nascent, concept for cost-effective grid-scale energy storage. While most studies report new active molecules and proof-of-concept prototypes, few discuss cell design. The direct translation of aqueous RFB design principles to nonaqueous systems is hampered by a lack of materials-specific knowledge, especially concerning the increased viscosities and decreased conductivities associated with nonaqueous electrolytes. To guide NAqRFB reactor design, recent techno-economic analyses have established an area specific resistance (ASR) target of <5 Ω cm². Here, we employ a state-of-the-art vanadium flow cell architecture, modified for compatibility with nonaqueous electrolytes, and a model ferrocene-based redox couple to investigate the feasibility of achieving this target ASR. We identify and minimize sources of resistive loss for various active species concentrations, electrolyte compositions, flow rates, separators, and electrode thicknesses via polarization and impedance spectroscopy, culminating in the demonstration of a cell ASR of ca. 1.7 Ω cm². Further, we validate performance scalability using dynamically similar cells with a ten-fold difference in active areas. This work demonstrates that, with appropriate cell engineering, low resistance nonaqueous reactors can be realized, providing promise for the cost-competitiveness of future NAqRFBs.

Grid-scale energy storage has emerged as a critical technology for alleviating the intermittency of renewables, improving the efficiency of the existing grid infrastructure, and offering frequency or voltage regulation. Redox flow batteries (RFBs) are particularly attractive storage devices for energy-intensive grid applications. In a typical RFB, charge-storing active species are dissolved in liquid electrolytes, which are housed in large and inexpensive tanks. The electrolytes are pumped through a power-generating electrochemical reactor, wherein the active species undergo reduction or oxidation on the surfaces of porous electrodes to charge or discharge the battery. This unique architecture offers several advantages over conventional battery systems, including decoupled power and energy scaling, long operational lifetimes, easy maintenance, simplified manufacturing, and high active-to-inactive materials ratio (particularly at long storage durations). Despite many attractive features, state-of-the-art RFBs are currently too expensive for widespread adoption. While the majority of RFB electrolytes are aqueous, transitioning to nonaqueous chemicals could enable higher cell potentials via wider electrolyte activities. Significant differences in the physicochemical and electrochemical properties of aqueous and nonaqueous electrolytes lead to uncertainty in how to effectively design flow cells for NAqRFBs. In particular, nonaqueous electrolytes can exhibit markedly different conductivities and viscosities, and active species for NAqRFBs typically exhibit faster reaction kinetics. AQFB development has recently benefited from a series of studies implementing a single electrolyte diagnostic flow cell technique to systematically evaluate cell performance. A schematic of this single electrolyte technique is provided in Figure 1a, where a flow cell is connected to a single electrolyte reservoir at 50% state-of-charge (SOC). The electrolyte passes through the positive side of the cell, where the active species are oxidized, and then loops back through the negative side of the cell, where the charged species are reduced. Since the same active species are present on both sides of the cell, crossover will not adversely impact cell lifetime or convolute data analysis. Additionally, the reservoir SOC is constant and effectively permits cell-level polarization and electrochemical impedance spectroscopy measurements at a steady-state. This technique most accurately mimics a RFB at 50%SOC when the conversion (f) through one side of the single electrolyte cell is low. The single pass conversion is described in Equation 1, where I is the total current (A), $C_{red}$ is the concentration of reduced species in the reservoir (mol m⁻³), n is the number of electrons transferred (-), F is the Faraday constant (C mol⁻¹), and Q is the electrolyte volumetric flow rate (m³ s⁻¹). Figure 1b illustrates how the electrolyte SOC will vary inside the flow cell as a function of normalized path length for various conversions.

$$ f = \frac{I}{C_{red}nFQ} $$

Single electrolyte studies rely on a stable active material; for example, prior AqRFB studies with this technique have exploited the
VO₂⁺/VO₃⁻ or V²⁺/V³⁺ redox couples. We previously reported 4-acetamido-(2,2,6,6-tetramethylpiperidin-1-yl)oxyl and its oxoammonium cation as a stable, model redox couple for studying NaqRFB electrolyte properties and cell performance. In that same report, we also postulated that the ferrocene / ferrocenium redox couple could be a viable candidate for diagnostic flow cell experiments if we also postulated that the ferrocene / ferrocenium redox couple, in conjunction with the single electrolyte technique, to demonstrate nonaqueous flow cell scalability, from a smaller cell (2.55 cm²) to a larger cell (25 cm²), without deviation of electrochemical performance. While the present study does not address all challenges associated with full RFBs, such as active species crossover or open circuit potential, these systematic experiments can guide future NaqRFB development and benchmark electrochemical performance.

**Experimental**

**Solution preparation.**—All solution preparation and electrochemical experiments were performed inside an argon (Airgas, AR UHP300, 99.999%) filled glove box (Inert Technologies, 4GB 2500, O₂ <5 ppm, H₂O <1 ppm) under ambient temperature (≈25°C). Propylene carbonate (PC, 99.99%) and acetonitrile (MeCN, 99.98%) solvents for electrochemical experiments were purchased from BASF and used as received. Lithium bis(trifluoromethane)sulfonimide (LiTFSI, 99.99%, BASF) and tetraethylammonium bis(trifluoromethane)sulfonimide (TEATFSI, 99%, lotitech) were used as received.

**Active species synthesis.**—N-(ferrocenylmethyl)-N,N-dimethyl-N-ethylammonium bromide ([Fc1N112⁺][Br⁻]) was synthesized, in the glove box, according to the following literature procedure (Figure 2). Dimethylaminomethyl)ferrocene (19.4 g, 79.8 mmol, 96%, Sigma Aldrich) was added to 150 mL of MeCN (Extra Dry 99.9%, Acros Organics). The flask was sealed with a rubber stopper, and the solution was stirred for 15 min. Bromoethane (6.2 mL, 83.0 mmol, 98%, Sigma Aldrich) was added dropwise at a rate of ≈0.5 mL min⁻¹. The solution was stirred overnight. The precipitate was filtered and washed with ≈25 mL of diethyl ether (99%, Alfa Aesar). Yield: 20.20 g (71.9%).

[Fc1N112⁺][TFSI⁻] was generated through an ion exchange in water (Figure 2). ([Fc1N112⁺][Br⁻] (12.3235 g, 35 mmol) was added to 130 mL of deionized water (DI H₂O). LiTFSI (13.36 g, 47 mmol) was dissolved in a separate flask with 50 mL of DI H₂O. The LiTFSI solution was added dropwise into the [Fc1N112⁺][Br⁻] solution in the round bottom flask and stirred for 1 h. The yellow [Fc1N112⁺][TFSI⁻] product (Figure 2, bottom left) was filtered and dried under vacuum for 24 h at room temperature. Yield: 24.72 g (97.9%).

[NiFc1N112⁺][TFSI⁻] was synthesized via a chemical oxidation and subsequent ion exchange (Figure 2). ([Fc1N112⁺][Br⁻] (12.3235 g, 35 mmol) was added to a 130 mL of DI H₂O. Iron(III) chloride hexahydrate (18.921 g, 70 mmol, 98%, Sigma Aldrich) was added to the solution and stirred for 1 h. LiTFSI (26.72 g, 93 mmol) was dissolved in a separate flask with 50 mL of DI H₂O. The LiTFSI solution was added dropwise into the [Fc1N112⁺][Br⁻] solution in the round bottom flask and stirred for 1 h. The yellow [Fc1N112⁺][TFSI⁻] product (Figure 2, bottom left) was filtered and dried under vacuum for 24 h at room temperature. Yield: 24.76 g (85.0%).

**Nuclear magnetic resonance spectroscopy (NMR).**—¹H NMR spectra were acquired using a Bruker Avance 400. Deuterated chloroform (D, 99.96%) was used as received from Cambridge Isotope Laboratories. NMR spectra were aligned using the residual solvent peak and reported versus tetramethylsilane (TMS). NMR spectra were obtained for only [Fc1N112⁺][TFSI⁻].

**Cyclic voltammetry and ultramicroelectrode.**—For both cyclic voltammetry (CV) and ultramicroelectrode (UME) measurements, three-electrode cells containing a 5 mL solution comprised of active species / 0.5 M TEATFSI / MeCN were used. The active species concentrations were 5 mM and 10 mM for CV and UME experiments, respectively. A coiled platinum wire (BASI) served as the counter electrode, and a Ag/AgCl reference electrode was used.

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**Figure 1.** (a) Single electrolyte flow cell configuration employing the Fc1N112⁻/²⁻ redox couple. (b) Schematic plot illustrating the electrolyte SOC as a function of path length for various reactant conversions.
electrode, and a fritted Ag/Ag⁺ quasi-reference electrode (BASi) was filled with silver tetrafluoroborate (0.1 M, 98%, Sigma Aldrich) in MeCN. The quasi-reference electrode was calibrated against the ferrocene / ferrocenium (Fc/Fc⁺) redox couple, in the supporting electrolyte of interest, and all voltammogram potentials are reported against the Fc/Fc⁺ reference calibration, in accordance with a prior literature procedure. CV measurements utilized a 3 mm diameter glassy carbon working electrode (BASi) and were collected using a Biologic VMP3 potentiostat with 100% solution resistance compensation (iR-correction). The 3 mm glassy carbon electrode was prepared by rinsing with DI H₂O, polishing with 5 μm alumina suspension, sonicking for 1 min, and then rinsing with DI H₂O a second time. The electrode was then dried before taking CV measurements. UME measurements implemented an 11 ± 2 μm carbon fiber working electrode (BASi) and were collected using a CH Instruments 630 potentiostat.

Conductivity and viscosity measurements.—Electrolyte conductivity measurements were collected using a two-electrode, Swagelok style conductivity cell and calibration method that have been previously described in literature. All electrolyte conductivity measurements were performed in triplicate. Separator conductivity measurements were also performed using a two-electrode, spring-loaded Teflon gaskets sealed the separators and electrodes into the cell; electrodes were compressed by 20 ± 2%, affording a final thickness of 304 ± 49 μm. By comparing the elastic moduli of the separators (≈400 MPa) and the carbon paper (≈4.4 MPa), we assume that, due to their significantly higher elastic moduli, the separators do not compress by any appreciable amount within the active area of the cell. Flow cells were assembled outside of the glove box and purged under vacuum (~91 kPag) three times before entering the inert atmosphere.

A sealed perfluoroalkoxy alkane (PFA) jar (10 mL, Savillex) served as the electrolyte reservoir. 10 or 30 mL of electrolyte was employed in the single electrolyte flow cell experiments for the 2.55 cm² and 25 cm² cells, respectively. A peristaltic pump (Masterflex L/S series) was used to drive electrolyte through the flow cell and reservoir. For the 2.55 cm² cell, PharMed BPT tubing (Masterflex L/S 14HP) was used inside a high-performance pump head and coupled to PFA tubing (1.6 mm inner diameter, Swagelok) that connected to the flow cell. For the 25 cm² cell, Norprene tubing (3.18 mm inner diameter, Masterflex L/S 16) was used inside an easy-load II pump head and connected directly to the flow cell.

Prior to polarization or impedance measurements, all cells were preconditioned by applying a constant potential of 0.4 V for 30 min, using an Arbin battery tester (FBTS-8). The preconditioning step allowed the cell to equilibrate, in accordance with prior single electrolyte flow cell studies. Additionally, since the single electrolyte configuration is employed (Figure 1a), the reservoir SOC does not change during the preconditioning step. Impedance measurements were collected with a 0.010 V sine wave about the open circuit potential across a frequency range of 1 MHz to 5 mHz using the Biologic VMP3 potentiostat. Potentiostatic measurements were performed to acquire polarization data from 0 to 0.6 V in 0.050 V increments using the Arbin battery tester. Potentiostatic holds were applied for 3 min, recording data every second, and the final polarization curves were computed as the mean of the last 30% of data points.

Due to the electronic resistance associated with the hermatically sealed cable connections through the glove box, the ohmic resistance contribution from the test leads was subtracted from all polarization and impedance measurements. To quantify the resistance contribution of the test leads for impedance data, the Biologic test leads were short...
circuited together, and an impedance measurement was collected to directly obtain the lead resistance of the impedance channel, which was \( \approx 0.05 \, \Omega \). This value was subtracted from the real contributions of all Nyquist plots. For polarization measurements, the Arbin test leads were short circuited together and their current-voltage characteristic was measured by applying galvanostatic holds in the range of 0–4 A. From the slope of the current-potential curve, the Arbin test leads through the glove box yielded a resistance of \( \approx 0.12 \, \Omega \). This resistance value was used to calculate an effective potential drop across the test leads during flow cell experiments by multiplying the lead resistance and the experimentally measured current. The computed test lead potential drop was subtracted from cell potential measurements.

Results

Active species voltammetry.—Voltammetric analysis is used to confirm both the facile redox kinetics associated with the Fc1N112\(^{+/2+}\) couple and the effectiveness of the chemical oxidation step. Figure 3a depicts CVs for the reduced species ([Fc1N112\(^+\)][TFSI\(^-\)]), and the initial scan direction is anodic (oxidative). Meanwhile, Figure 3b depicts CVs for the oxidized species ([Fc1N112\(^{2+}\)][TFSI\(^-\)]), and the initial scan direction is cathodic (reductive). We analyze the reduced and oxidized species separately with CV to confirm that Fc1N112\(^{2+}\) maintains redox activity and facile reaction kinetics after the chemical oxidation (Figure 2). Figure 3a and Figure 3b illustrate that the CV (cycle 2) peak separation is unchanging with scan rate for Fc1N112\(^{+/2+}\) in both oxidation states. The peak-to-peak separation across all CV scans, for both the reduced and oxidized compounds, is 0.067 \( \pm 0.001 \) V, which is slightly larger than the idealized separation (0.059 V), likely due to incomplete IR-correction. The peak-height ratio is 1.04 \( \pm 0.009 \) across all scans, for both compounds. The peak-height ratio is calculated by first subtracting the background current, according to a prior literature procedure, to acquire corrected peak heights and then dividing the corrected peak scan current of the backward scan by the corrected peak current of the forward scan. Finally, the reduced and oxidized species exhibit different iron oxidation states and associate with a different number of TFSI\(^-\) anions. Thus, each species is likely to exhibit a different solvated radius and subsequently different diffusion coefficient, which will impact the observed peak currents and explains the asymmetry between Figure 3a and Figure 3b.

Figure 3c and Figure 3d show UME scans for Fc1N112\(^+\) and Fc1N112\(^{2+}\), respectively. Note that for Fc1N112\(^+\), only an anodic (oxidizing) current is present, whereas for Fc1N112\(^{2+}\), only a cathodic (reducing) current is present. Hence, the UME results indicate that the Fc1N112\(^+\) sample contains only the reduced active species and the Fc1N112\(^{2+}\) sample contains only the oxidized active species, at least to the detection limit of this method. Confirming that the reduced and oxidized species are isolated lends confidence to the SOC (50%) of the solutions prepared for full cell measurements. Further, the magnitude of the steady-state currents from UME measurements are different for the reduced and oxidized species, supporting the prior hypothesis that the asymmetry in the CV data (Figure 3a and Figure 3b) is caused by diffusion coefficients that vary with active species oxidation state. A parallel data set to Figure 3 found the same electrochemical reversibility and oxidation states in a supporting electrolyte of 0.5 M LiTFSI / PC, but is not presented here for brevity.

Separator and electrolyte properties.—Nonaqueous electrolytes encompass a broad spectrum of materials and properties, where electrolyte conductivities and viscosities can easily vary by an order of magnitude. While such a range of materials and properties provides exciting design possibilities for full NAqRFBs, generalizing performance limitations becomes difficult. To capture the breadth of available electrolyte properties, we identify two supporting electrolytes, LiTFSI / PC and TEATFSI / MeCN, which eivince an approximately
Table I. Conductivities and viscosities of electrolytes employed in diagnostic flow cell experiments.

| Fc1N112/2+ Conc. (M) | Salt (0.5 M) | Conductivity (mS cm⁻¹) | Viscosity (mPa·s) |
|-----------------------|--------------|-------------------------|------------------|
| 0.25                  | TEATFSI MeCN | 30.8 ± 0.5              | 0.7 ± 0.1        |
| 0.5                   | TEATFSI MeCN | 27.8 ± 0.9              | 1.1 ± 0.1        |
| 1.0                   | TEATFSI MeCN | 16.9 ± 1.1              | 3.7 ± 0.1        |
| 1.0                   | LiTFSI PC    | 2.8 ± 0.4               | 33.2 ± 0.1       |

change for different electrolytes, even when the separator morphology is the same. This discrepancy is especially large when comparing Celgard 2500 and 3501, which have the same mechanical properties, but different surface coatings. The disagreement in s/t indicates that separator wetting plays a significant role in determining R_mem. We also present the experimentally determined ratio of electrolyte-to-separator conductivity (κ/κ_eff, MacMullin number), which is ≈2x higher for Celgard as compared to Daramic, and offers another descriptor of the effect of electrolyte conductivity on separator conductivity. The MacMullin numbers for Celgard are in good agreement with a prior report.

\[ R_{mem} = \frac{t}{ek} \]

Celgard and Daramic microporous separators do not exhibit sufficiently high selectivity for practical NaAgRFB applications, but the single electrolyte configuration (Figure 1a) permits flow cell studies without concerns of crossover degrading performance. Generally, as the separator thickness increases (i.e., Celgard to Daramic) or the electrolyte conductivity decreases, R_mem will increase. For example, when switching from Daramic 175 to Celgard 3501, with the PC-based electrolyte and 1 M actives, the R_mem drops by ≈4.4×. Similarly, switching from the PC- to MeCN-based electrolyte, with Daramic 175 and 1 M actives, the R_mem reduces by ≈6.6×. Further, the MeCN-based electrolytes where the separator ASR rises with increasing active species concentration, mirroring the decrease in electrolyte conductivity quantified in Table I. Thus, the matrix of available separators and electrolyte compositions allows us to study flow cell performance with a variety of effective separator ASR values (Table II).

Separator and supporting electrolyte comparison.—Increasing separator conductivity is arguably the most direct method for improving RFB reactor performance because a decrease in separator resistance will lead to an equivalent reduction in total cell ASR. Figure 4a illustrates this effect by comparing polarization of cells using either Celgard or Daramic separators with 1 M Fc1N112/2+. For both the PC- and MeCN-based electrolytes, the Celgard separators yield improved cell performance over their Daramic counterparts due to lower separator resistance. The Nyquist plots in Figure 4b and Figure 4c illustrate that, for a fixed electrolyte composition, the separator choice drastically affects the high frequency intercept (R_Ω), with no other major changes to the Nyquist plots. These results indicate that varying the separator type directly affects the ohmic contribution to the total cell ASR, and the impedance reductions observed in Figure 4b and Figure 4c are equivalent to the separator ASR reductions in Table II, when switching from Daramic to Celgard. Additionally, the high frequency intercepts are only slightly larger than the corresponding values of R_mem, indicating that the R_mem dominates the total ohmic contribution to the cell resistance; for example, the cell with a Daramic separator and 0.5 M Fc1N112/2+ / 0.5 M TEATFSI / MeCN electrolyte has R_Ω = 1.41 Ω cm² and R_mem = 1.27 Ω cm².

Beyond the separator effect, Figure 4a also indicates that the MeCN-based electrolyte always outperforms the PC-based electrolyte, regardless of the separator thickness or identity. First, this performance enhancement is partially due to improved separator conductivity with the MeCN-based electrolyte (Table II). Comparing the
electrolytes and a Celgard separator. By subtracting $R_\Omega$ from both plots (Figure 4d), the variation in the diameters of the two Nyquist plots becomes apparent. The Nyquist plot for the cell with MeCN-based electrolyte exhibits a semicircle with $\approx 4 \times$ smaller diameter than the PC-based electrolyte, which is due to the decreased mass transfer resistance associated with the active species transport to the electrode surface, as will be elucidated in a following flow rate dependence study. The MeCN-based electrolyte exhibits improved mass transfer rates due to its lower viscosity as compared to the PC-based electrolyte (Table 1). Consideration of electrolyte viscosity as an electrochemical performance parameter, independent of conductivity, is a critical design concern for RFBs, though of lesser concern for non-flowing electrochemical cells (e.g., lithium ion batteries). Since RFBs rely on convection as the primary form of active species mass transfer, the electrolyte viscosity will directly affect reactant delivery rates within the porous electrode. Thus, electrolyte design for NaAqRFBs must consider viscosity as a key materials optimization parameter, and, in the case of this work, MeCN vastly outperforms PC as a base solvent for a low viscosity and high conductivity electrolyte.

**Flow rate effects.**—The impedance variations from Figure 4d hint that mass transfer rates are critical in determining the total ASR for RFBs, but only a handful of prior reports have systematically studied mass transfer effects in AqRFBs, with no reports directly relating to NaAqRFBs. To begin addressing this knowledge gap, Figure 5 highlights nonaqueous flow cell performance for the PC- and MeCN-based electrolytes, with a Daramic separator, at 4 flow rates spanning greater than an order of magnitude. As anticipated, for both electrolytes, increasing the electrolyte flow rate improves overall cell performance: higher current densities are achieved at a fixed cell potential (Figure 5a and Figure 5b). Considering the Nyquist plots in Figure 5c and Figure 5d, the low frequency intercept ($R_{DC}$) decreases with increasing flow rate, and no other major changes are observed in the overall shape or $R_\Omega$. Note in Figure 5c that the variations in $R_\Omega$ (approximately $\pm 1 \Omega \text{cm}^2$) are small in comparison to the total cell ASR ($> 21 \Omega \text{cm}^2$), representing <5% variation, which is believed to be within experimental error. Thus, the Nyquist plots validate that modifying flow rate affects only the mass transfer resistance. For both electrolyte systems, the electrochemical performance of the flow cell exhibits a diminishing rate of return with increasing electrolyte flow rate. For example, consider Figure 5b, where the improvement in performance from $1-5 \text{ mL min}^{-1}$ is much larger than the improvement from $5-10 \text{ mL min}^{-1}$. We hypothesize that, as the flow rate increases, the cell approaches the limit of infinitely fast mass transfer; typically, mass transfer coefficients in porous media increase with flow velocity to a power $\leq 1.6$. Thus, in the limit of high electrolyte velocity, the ohmic and charge transfer losses dominate the cell’s resistive characteristics. As such, selecting an optimal flow rate will require balancing the cell ASR with pumping losses; beyond a certain critical flow rate, the ASR reduction will be smaller than the magnitude of pumping loss required to boost the flow rate. One study has attempted this optimization for NaAqRFBs, with several other examples for AqRFBs.

While both electrolyte systems exhibit a flow rate dependence, the cell with a PC-based electrolyte exhibits performance that is less sensitive to flow rate variations than the cell with MeCN. Figure 5c highlights that $R_\Omega$ is much larger for the PC-based electrolyte cell than all other real impedance contributions (i.e., the diameter of the Nyquist plots), and therefore, changes in the mass transfer resistance due to changes in electrolyte flow rate only account for a small fraction of the total cell ASR. By contrast, the MeCN-based electrolyte cell has a $R_\Omega$ that is on the same order of magnitude as all other resistance contributions ($R_\Omega \approx (R_{DC} - R_{R_z})$), so variations in mass transfer resistance account for a larger fraction of the total cell ASR. Thus, we can now describe a general trend for RFBs that as the separator conductivity decreases, cell performance will become more sensitive to variations in electrolyte flow rate.

**Active species concentration.**—Continuing to investigate mass transfer effects, Figure 6 shows polarization curves at varying active
Figure 5. Cell performance with varying electrolyte flow rates, containing 1 M Fe\text{II}/Fe\text{III} (50% SOC) and Daramic separators. (a) Polarization of 0.5 M LiTFSI / PC. Inset: Expanded dataset in current density range of 0–30 mA cm$^{-2}$. (b) Polarization of 0.5 M TEATFSI / MeCN. (c) Nyquist plots of 0.5 M LiTFSI / PC. (d) Nyquist plots of 0.5 M TEATFSI / MeCN.

Figure 6. Cell polarization with varying active species concentrations, implementing the Daramic / 0.5 M TEATFSI / MeCN configuration, for 4 different flow rates: (a) 0.5, (b) 1.0, (c) 5.0, and (d) 10 mL min$^{-1}$. 
species concentrations and flow rates for the MeCN-based electrolyte. At the lowest flow rate (0.5 mL min⁻¹, Figure 6a), the cell achieves limiting current density, which increases with increasing active species concentration. In fact, all current densities, at a fixed cell potential, increase with active species concentration. These two trends are anticipated as the cell with the highest active species concentration should exhibit the smallest mass transfer resistances, assuming all other cell parameters are held constant. As the flow rate increases, however, this trend no longer holds true; the 1.0 M electrolyte (highest active species concentration) performs worse than the lower concentration electrolytes (Figure 5c and Figure 6d).

To begin deconvoluting the unexpected cell performance trends at high active species concentrations, consider the Nyquist plots in Figure 7. Column i in Figure 7 shows Nyquist plots shifted by R₂ to various active species concentrations for the 4 flow rates depicted in Figure 6. At lower flow rates (e.g., 0.5 mL min⁻¹, Figure 7a,i), the diameters of the Nyquist plots decrease with increasing active species concentration. This anticipated trend is again due to decreasing mass transfer resistance with increasing active species concentration. However, as the flow rate becomes large (e.g., 10 mL min⁻¹, Figure 7d,i), the semicircles converge, regardless of the active species concentration, indicating that at sufficiently high flow rates the mass transfer resistance becomes insensitive to variations in active concentrations.

The lack of sensitivity to active concentrations in the Nyquist plots at high flow rates is an interesting result, but does not reconcile the behavior observed in Figure 6. Consider now the Nyquist plots in Column ii of Figure 7, which contain the entire impedance spectra. Note first that R₂ is different for all active species concentrations, with the 1.0 M electrolyte exhibiting the highest value of R₂. The experimentally measured electrolyte and separator conductivities (Table II) indicate that the 1.0 M electrolyte cell should exhibit the highest value of R₂ due to the higher resistance associated with that electrolyte system. At low flow rates, where mass transfer is the dominant resistive feature, R₂ decreases with increasing active species concentration, despite differences in R₂ (e.g., Figure 7a,ii). As flow rate increases and mass transfer rates improve, ohmic losses become the major resistance contribution and dominate the overall cell performance. Hence, variations in R₂ control Rₘₐₜ under conditions with facile mass transfer. This result illustrates the importance of considering all resistance contributions to R₂ so that the dominant resistive features can be identified and addressed appropriately.

An optimal active species exists that will balance the electrolyte conductivity, viscosity, and concentration to deliver the best overall electrochemical performance (lowest ASR) and does not necessarily correspond to the highest active species concentration possible. The cell ASR, however, is only one performance metric in a complex energy storage system, and while the ASR plays a significant role in determining the electrochemical stack cost, the active species concentration directly affects the electrolyte cost contribution to the total RFB cost. In brief, higher energy density electrolytes suppress solvent and tank costs, yielding lower chemical costs per unit energy stored, which is especially true for NaqRFBs where the solvent cost is expected to be an order of magnitude greater than that of a typical aqueous electrolyte. The decrease in electrolyte cost contribution competes with an increase in the reactor cost contribution associated with higher ASR. These two cost components must be balanced when selecting an optimal operating concentration.

Electrode thickness.—Several prior studies on AqRFBs have included electrode thickness optimization to deliver the best electrochemical performance for a particular electrolyte system. Increasing the electrode thickness increases ohmic losses through the porous electrode due to longer electron and ion path lengths to the current collector. However, increasing the electrode thickness also increases the available surface area for electrochemical reactions, which permits a larger exchange current and reduces charge transfer losses. Balancing these two effects, to optimize the total electrode resistance, becomes particularly important for RFBs with slow reaction kinetics. Finally, increasing the electrode thickness will reduce pressure drop for a fixed flow rate or lower intra-electrode velocity for a fixed pressure drop. In the latter case, mass transport resistances will increase with increasing electrode thickness.

To evaluate the sensitivity of NaqRFB performance to electrode thickness at equal compression, polarization and impedance analyses are performed on cells with thick (6×) and thin (2×) stacked carbon paper electrodes (Figure 8). For both a fixed intra-electrode velocity over the IDFF rib (Figure 8a) and fixed electrolyte flow rate (Figure 8b), the thinner electrode yields better overall electrochemical performance, but the difference is much smaller for the fixed intra-electrode velocity case (Figure 8a). When considering a flow-through porous electrode with fixed intra-electrode velocity, the electrochemical performance should be identical. Differences in the polarization curves in Figure 8a are likely due to differences in the specific velocity field created by the IDFF, which will vary nonlinearly with increasing thickness. Figure 8c supports this claim, as the different electrode thicknesses yield similarly shaped Nyquist plots. For a fixed electrolyte flow rate, the intra-electrode velocity decreases with increasing electrode thickness, yielding lower overall performance (Figure 8b) and much larger mass transfer losses, as indicated by a larger diameter Nyquist plot (Figure 8c).

While the mass transfer effects vary significantly for the fixed velocity and flow rate cases, the shape and location of the high-frequency intercept is unchanging, regardless of the electrode or flow condition. We hypothesize that for nonaqueous flow cells, the ohmic and charge transfer properties of the cell are insensitive to variations in electrode thickness, as compared to aqueous systems, for two reasons. First, the separator resistance for a NaqRFB is much greater than that of an AqRFB and dominates R₂. As such, variations in R₂ with increasing electrode thickness are small by comparison and negligibly affect the overall cell performance. Second, most active species for NaqRFBs, including FeC₁₁N₁₂₂, exhibit rapid reaction kinetics and thereby large exchange current densities. For example, ferrocene in nonaqueous electrolyte has been reported to have a kinetic rate constant (k₀) ~10⁻⁸ cm s⁻¹, which is two orders of magnitude greater than that of aqueous all-vanadium (k₀ ~10⁻³ cm s⁻¹). As an additional comparison, (2,2,6,6-tetramethylpiperidin-1-yl)oxyl (TEMPO), an organic redox active species considered for NaqRFBs, exhibits even faster rate constants: 1.0 × 10⁻⁴ cm s⁻¹, which is two orders of magnitude greater than that of aqueous all-vanadium for a typical carbon paper electrode. Flow cell ASR becomes sensitive to charge transfer losses for k₀ < 10⁻³ cm s⁻¹. Due to a lack of sensitivity to the ohmic and kinetic effects, within the practical range of thicknesses studied, the electrode thickness for NaqRFBs should be used as a parameter to balance mass transport rates with pressure drop.

Reactor scaling.—As previously mentioned, to date, most NaqRFB studies have not leveraged advanced aqueous flow cell designs, and, to the best of our knowledge, the largest nonaqueous flow cell reported had an active area of only 10 cm². While RFBs are scalable devices, the scaling relationships are not necessarily straightforward (e.g., matching pressure drop) and scale-dependent factors exist (e.g., edge effects) that can impact performance. The 2.55 cm² cell design, used to collect all the data shown to this point, has practical value as only relatively small quantities of active materials are required for cell testing. Experiments with the 2.55 cm² cell illustrate a range of performance tradeoffs, but, to ensure experimental validity, we must demonstrate performance scalability, at least to the size of more typical aqueous RFB studies. To this end, we constructed a 25 cm² cell to specifically validate observations in our small reactor and to generally evaluate performance scalability.

The 2.55 cm² and 25 cm² cells are designed to yield the same pressure drop and electrochemical performance for a fixed area specific flow rate. Darling and Perry described an analytical framework for calculating the pressure drop through an IDFF for flow batteries. The original derivation by Owejan et al. was targeted towards...
Figure 7. Nyquist plots of cells with varying Fc1N112+/2+ (50% SOC) concentrations, implementing the Daramic / 0.5 M TEATFSI / MeCN configuration, for 4 different flow rates: (a) 0.5, (b) 1.0, (c) 5.0, and (d) 10 mL min⁻¹. Column (i) presents the real axis shifted by $R_O$, whereas column (ii) presents the total real resistance.

Figure 8. Cell performance with varying electrode thicknesses, containing 1 M Fc1N112+/2+ (50% SOC), Celgard 2500, and 0.5 M TEATFSI / MeCN. (a) Polarization at fixed internal electrode velocity. (b) Polarization at fixed electrolyte flow rate. (c) Nyquist plots at fixed internal electrode velocity. (d) Nyquist plots at fixed electrolyte flow rate.
polymer-electrolyte fuel cells and is available in Ref. 77. The pressure drop through the IDFF ($\Delta P$) can be computed as shown in Equation 3, where $\Delta P_{ch}$ (Pa) is the pressure drop through the rectangular flow field channels, and $\zeta$ is a dimensionless geometric factor (-).

$$\Delta P = \Delta P_{ch} \left( 1 + \frac{2 + 2 \cosh \zeta}{\zeta \sinh \zeta} \right)$$ \[3\]

The pressure drop through the rectangular channel of the IDFF is calculated from the analytical expression in Equation 4, where $\mu$ is the electrolyte viscosity (Pa·s), $L_{ch}$ is the channel length (m), $d_h$ is the hydraulic diameter of the channel (m), and $v_{ch}$ is the electrolyte velocity in the channel (m s$^{-1}$).

$$\Delta P_{ch} = \frac{32 \mu v_{ch} L_{ch}}{d_h^2}$$ \[4\]

The channel velocity is defined in Equation 5, where $Q$ is the volumetric electrolyte flow rate (m$^3$ s$^{-1}$), $N$ is the number of channels (-), $w$ is the channel width (m), and $h$ is the channel height (m).

$$v_{ch} = \frac{Q}{N w h}$$ \[5\]

The geometric factor $\zeta$, which appears in Equation 3, is calculated according to Equation 6, where $k$ is the electrode permeability (m$^2$), $L_e$ is the electrode thickness (m), and $S$ is the path length of the electrolyte through the electrode and over the rib (m).

$$\zeta^2 = \frac{128 L_{ch}^2 k L_e}{S d_h^2 w h}$$ \[6\]

While the mean fluid path length through the IDFF electrode can be computed numerically,72 such a calculation is beyond the scope of this work. We estimate $S$ as the sum of the electrode thickness and width of two channels, as shown in Equation 7.

$$S \approx 2w + L_e$$ \[7\]

To match the pressure drop between two IDFFs of differing size or geometry, the equality relationship presented in Equation 8 must hold.

$$\frac{Q_1 L_{ch,1} N_1 w_1 h_1 d_h^2_{1,2}}{Q_2 L_{ch,2} N_2 w_1 h_1 d_h^2_{2,1}} = \left( \frac{1 + 2 \cosh (\zeta_{1,2} / 2)}{1 + 2 \cosh (\zeta_{2,1} / 2)} \right)$$ \[8\]

Table III carries out a calculation of the pressure drop through the IDFF for a fixed area specific flow rate, $Q/A = 6.5 \times 10^{-3}$ cm s$^{-1}$, for both the 2.55 cm$^2$ and 25 cm$^2$ cells, which develop near identical pressure drops of 20.0 kPa and 22.0 kPa, respectively. Table III also lists all relevant parameters for the pressure drop calculation.

Figure 9 compares the electrochemical performance of the 2.55 cm$^2$ and 25 cm$^2$ cells, at two different area specific flow rates ($Q/A$). Note that for cell polarization (Figure 9a), measurements on the 25 cm$^2$ cells do not extend to the same current densities as the 2.55 cm$^2$ cell due to larger ohmic loss through the Arbin battery tester cables. Similarly, the two area specific flow rates are selected to remain within the calibrated flow rate range of the peristaltic pump for both flow cell configurations. Figure 9a clearly indicates scalable electrochemical performance for the area specific flow rates and current densities selected, at both flow conditions, as the polarization curves overlay.

Table III. Relevant flow field geometry parameters for computing pressure drop through the 2.55 cm$^2$ and 25 cm$^2$ IDFFs. An example calculation is carried out for a fixed area specific flow rate: $Q/A = 6.5 \times 10^{-3}$ cm s$^{-1}$.

| Parameter | Description | Units | 2.55 cm$^2$ Value | 25 cm$^2$ Value |
|-----------|-------------|-------|------------------|-----------------|
| $d_h$     | Hydraulic Diameter | m     | $6.67 \times 10^{-4}$ | $6.67 \times 10^{-4}$ |
| $h$       | Channel Height   | m     | 0.0005           | 0.0005          |
| $k$       | Electrode Permeability | m$^2$ | $10^{-12}$ | $10^{-12}$ |
| $L_{ch}$  | Channel Length   | m     | 0.0150           | 3 $\times 10^{-4}$ |
| $L_e$     | Electrode Thickness | m    | $3 \times 10^{-4}$ | $3 \times 10^{-4}$ |
| $\mu$     | Electrolyte Viscosity | Pa·s | 0.001            | 0.001           |
| $N$       | Number of Inlet Channels | -    | 4                | 12              |
| $\Delta P$ | Open Channel Pressure Drop | Pa | 54.9        | 511             |
| $Q$       | Flow Rate        | m$^3$ s$^{-1}$ | $1.67 \times 10^{-7}$ | $1.63 \times 10^{-6}$ |
| $S$       | Flow Path Length | m     | 0.00230          | 0.00230         |
| $v_{ch}$  | Channel Velocity | m s$^{-1}$ | 0.084           | 0.272           |
| $w$       | Channel Width     | m     | 0.001            | 0.001           |
| $\zeta$   | Geometric Factor  | -     | 0.139            | 0.433           |
| $\Delta P$ | IDFF Pressure Drop | Pa | $2.00 \times 10^5$ | $2.20 \times 10^4$ |
of alkaline or neutral AqRFBs. A recent modeling study of ohmic-limited NAqRFBs, in the limit of fast mass transport at 100% SOC, suggested that the lowest possible ASR would be \( R_{\Omega} \approx 3.4 \, \Omega \text{ cm}^2 \) (\( \approx 89 \, \text{mA cm}^{-2} \) at 0.3 V overpotential); our experimental work, incorporating realistic mass transfer at 50% SOC, exceeds this predicted performance by >40% for overpotentials <0.3 V.

While the excellent electrochemical performance in Figure 10 demonstrates the potential for low ASR nonaqueous flow cells, technical hurdles remain. The implementation of the MeCN-based electrolyte and Celgard 2500 separator is critical in achieving low ASR, but the Celgard 2500 separator is impractical for a NAqRFB device since it offers no selectivity for small redox active molecules. Implementing Celgard 2500 in full flow cell would require mixed active species electrolytes, which would be cost prohibitive or emerging large polymeric active species, which may yield high viscosity electrolytes with poor mass transfer characteristics. Additionally, the highly soluble Fc1N112 model active species and low viscosity MeCN-based electrolyte facilitates small mass transfer resistances. Discovery and synthesis of stable and soluble active species for NAqRFBs is challenging, and quantifying variations in mass transfer rates for electrolytes of varying viscosity has yet to be reported. Simultaneously tailoring active species radius and separator pore size offers a promising strategy to enabling NAqRFBs with sufficiently low ASR, high selectivity, and good mass transport. Further, while RFBs typically exhibit improved safety over other battery types (e.g., lithium-ion, sodium-sulfur), the safety associated with the implementation of nonaqueous solvents in RFBs must be investigated as the technology continues to advance. Ultimately, the performance depicted in Figure 10 sets a benchmark for the electrochemical performance of nonaqueous flow cells, however, numerous other criteria must be simultaneously met (i.e., cell potential, stable actives) to achieve economically viable battery designs.

**Conclusions**

A major criticism of NAqRFBs has been an inability to achieve power densities comparable to their aqueous counterparts. Several studies have attempted to boost the cell potential of NAqRFBs through the discovery and development of new active materials, which offer one pathway toward improving power density, reducing reactor costs, and, consequently, decreasing the overall battery cost. The purpose of this study is to demonstrate the achievable current densities in high performance nonaqueous flow cells, which can eventually be coupled with a high voltage redox chemistry to produce a full cell with improved power density. This systematic study of resistive losses in NAqRFBs culminates in the demonstration of ASR values as low as 1.7 \( \Omega \) cm\(^2\), along with current densities near 150 mA cm\(^{-2}\) at 0.3 V overpotential (Figure 10). These metrics exceed the recommended performance for economically feasible nonaqueous flow cells. This study is also the first to illustrate scalability for nonaqueous flow cell design, highlighting identical polarization performance over a 10x active area increase (Figure 9a).

The performance achieved in this study is unprecedented in the NAqRFB literature and stems from a logical analysis of the key resistive features in a nonaqueous flow cell and subsequent methodology to reduce those resistive losses. First, ohmic losses are the largest hurdle to overcome, due to the relatively low conductivities of nonaqueous electrolytes, as compared to aqueous electrolytes, and the subsequently low separator / membrane conductivities. Identifying thinner or more conductive separators can alleviate ohmic resistances; recent literature on size selective separators for NAqRFBs is providing a pathway toward separators that are sufficiently conductive and selective. Interestingly, the ohmic loss through the porous electrode is negligible compared to the ohmic loss through the separator. Mass transfer losses are the second largest impediment to flow cell performance and can be alleviated by increasing electrolyte flow rate, increasing active species concentration, or decreasing electrolyte viscosity. Finally, charge transfer losses are negligible for most active species investigated for NAqRFBs because of the relatively fast
reaction rate constants associated with redox active species in nonaqueous electrolytes, as compared to those in aqueous electrolytes. Our results contrast the major resistive losses and cell design sensitivity of historic aqueous RFBs (e.g., all-vanadium), which typically exhibit significant ASR contributions from charge transfer due to typically low reaction rate constants ($k_0 < 10^{-3}$ cm$^{-1}$ s$^{-1}$). Combining facile kinetics with a relatively small ohmic resistance from the porous electrode indicates that nonaqueous flow cell performance is relatively insensitive to variations in electrode thickness, which is not the case for typical aqueous RFBs. This work highlights the promising performance of nonaqueous flow cells, but this performance must be coupled with a sufficiently selective separator and stable, high-voltage ($>3$ V) chemistry. Moving forward, our work will focus on leveraging the ASR reduction principles outlined here to inform the design and development of more promising NaRFB separators and electrolytes.

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