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Computational and Experimental Study of Convection in a Vanadium Redox Flow Battery Strip Cell Architecture

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Abstract: The impact of convection on electrochemical performance, performance distribution, and local pressure drop is investigated via simple strip cell architecture, a cell with a single straight channel. Various channel depths (0.25, 0.5, 1, 2.5 mm) and flow rates ($10\text{--}50\text{ mL min}^{-1}\text{ cm}^{-2}$) are employed to induce a wide range of electrolyte velocities within the channel and electrode. Computational flow simulation is utilized to assess velocity and pressure distributions; experimentally measured in situ current distribution is quantified for the cell. Although the total current in the cell is directly proportional to electrolyte velocity in the electrode, there is no correlation detected between electrolyte velocity in the channel and the total current. It is found that the maximum achievable current is limited by diffusion mass transport resistance between the liquid electrolyte and the electrode surfaces at the pore level. Low electrolyte velocity induces large current gradients from inlet to outlet; conversely, high electrolyte velocity exhibits relatively uniform current distribution down the channel. Large current gradients are attributed to local concentration depletion in the electrode since the velocity distribution down the channel is uniform. Shallow channel configurations are observed to successfully compromise between convective flow in the electrode and the overall pressure drop.

Keywords: vanadium redox flow battery; strip cell; convective mass transport; current distribution; CFD; model validation

1. Introduction

The National Renewable Energy Laboratory (NREL) estimates that renewable energy sources (solar, wind power and hydropower) could be responsible of 70–75% total electricity generation in the United States by 2050 [1]. Due to the intermittent nature of renewable energies, large-scale energy storage technologies are indispensable for high penetration of variable electricity generation to the power grid. Energy storage technologies can address the challenges: electricity service stability, flexibility, reliability, and resilience on the power grid. Among energy storage systems, vanadium redox flow batteries (VRFBs) have been pursued due to their flexibility and scalability, high coulombic efficiency, and long cycle life. However, widespread commercialization of VRFBs suffers from high system capital cost. The US Department of Energy (DOE) proposed a target system capital cost under 150 USD/kWh for commercialization of energy storage systems [2]. To achieve this goal, VRFB cell component costs (membranes and chemicals), which dominate overall system costs, need to be reduced [3,4]. Improving electrolyte utilization and overall system efficiency are the most viable approaches to reduce overall system costs. Recent efforts include increasing solubility of the four

vanadium species in the solvent [5–8], enhancing separators to be more ionically conductive, alleviating crossover [9–17], facilitating electrochemical kinetics [18–21], improving mass transport, and reducing parasitic pump losses [22–25].

In VRFB cells, transport of active species to/from electrode reaction surfaces is critically important. Insufficient active species transport leads to concentration overpotential, also known as mass transport polarization, due to reactant depletion and/or product accumulation at electrode surfaces. Increased overpotential results in decreased voltage efficiency, reduced accessible state of charge (SOC), and reduced effective energy capacity of the battery. One of the most straightforward approaches to mitigate mass transport losses is increasing vanadium concentration in the solvent. It has been shown that vanadium concentration in the solvent can be increased (maximum 3 M vanadium electrolyte was achieved) by using mixed acid solution [5] and additives [6]. Despite these improvements, vanadium species solubility in the solvent is still limited [26]. Increasing the flow rate is another simple approach frequently found to enhance convective mass transport in the electrode [27–32]. However, due to parasitic pumping losses, increasing the flow rate becomes an optimization problem highly sensitive to other system parameters. Common flow field designs, including flow-through (aspect ratio and equal path length) and flow-by (e.g., parallel, interdigitated and serpentine) have been widely investigated in VRFB literature [22,23,33–41]. It has been also reported that channel geometry (channel height, channel width, channel length, and land or rib width) influence electrochemical performance [36,39,42,43]. In addition to flow field impacts, the electrode is one of the most critical components since electrochemical reactions occur on the electrode surface. Many studies have attempted to improve ion transport by modifying electrode structure (thickness, porosity, and tortuosity) [35,44–48]. However, there is a sensitive balance between permeability and electrochemical surface area in the electrode; in general, higher permeability is achieved at the cost of reduced active surface area.

Various in situ distributed diagnostic techniques have been utilized to understand underlying mechanisms in VRFBs [11,49–52]. Clement et al. employed in-plane current distribution measurement to investigate mass transport for a range of cell and operating parameters [53]. Houser and co-workers revealed contributions from the two dominating mass transport mechanisms by comparing current distributions for interdigitated and serpentine flow field designs [22]. In our previous study [54,55], current distribution was implemented for a single channel (“strip cell”) architecture to isolate diffusion and convection mass transport mechanisms. Enhanced electrochemical performance and relatively uniform current density distribution were attributed to increased convective flow in the porous electrode [56]. However, the VRFB is a complex system with multiple interrelated parameters (e.g., mass transport and electrochemical reactions) affecting electrochemical cell performance; experimentally disentangling these interrelated parameters is very challenging. Additionally, while experimental investigation often provides quantitative physicochemical measurements of VRFB behavior, it is relatively costly, slow, and limited by available equipment. Computational analysis through first-principles based modeling is a complementary approach with unique benefits discussed in the following paragraphs. Thus, a comprehensive and properly validated computational simulation is essential to understand physicochemical processes in VRFBs.

Among VRFB models, cell-level modeling, specifically via computational fluid dynamics (CFD), is the most common in literature to date, in comparison with stack-scale, pore-scale, and electrode modelling. In general, the simulation domain includes porous electrodes and flow plates. Continuum equations include conservation of mass and momentum to simulate electrolyte distribution in the VRFB cell [36]. Maximum electrolyte utilization and uniform current density distribution are directly influenced by electrolyte distribution in VRFBs; this distribution is highly dependent on electrolyte flow behavior at the transition between flow field channels and electrode porous media. Ke et al. has shown that the limiting current can be predicted by calculating the electrolyte penetration to the electrode: increased convection in the porous layer yielded improved performance [37,40,57]. Houser et al. achieved higher electrode velocity via equal path length (EPL) and aspect ratio (AR) flow

field designs that show superior electrochemical performance [23]. Maurya et al. investigated different flow field designs where enhanced electrochemical performance was attributed to the increased electrode flow velocity [41]. More sophisticated models that include electrochemical reactions have also been developed to explore electrolyte velocity impact. You et al. investigated the correlation between mass transfer coefficient and electrolyte velocity by measuring limiting currents [58]. Kok et al. extensively studied the impact of electrode morphology and cell architecture on electrochemical performance. Increased electrolyte velocity in the electrode improves mass transport losses by reducing diffusion path length [45]. Other works have also reported the convection impact on electrochemical performance [24,33,39,59,60]. However, these studies used relatively complex flow fields (e.g., parallel, interdigitated, serpentine) or employed overall cell performance analyses (e.g., polarization curve, limiting current). Considering that the local electrolyte velocity, along with local pressure drop, vary along the channel, distributed diagnostic measurement is essential to understand the impact of convection on VRFB electrochemical performance.

The goal in this study is to more directly measure the impact of convection on VRFB electrochemical performance. To achieve this, a computational fluid dynamics (CFD) model using COMSOL Multiphysics® software along with experimental, in situ, localized current distribution diagnostics are utilized. Pressure drop analysis is also employed to support the conclusions and partially validate the modeling results. The CFD analysis used in this study reveals key hydrodynamic relations both in the channel and electrode so that the convection transport mechanism and its impact on electrochemical performance (both overall and distributed) is investigated more fundamentally. Experiments and numerical simulations were conducted for a strip cell architecture which has a simplified geometry with one straight channel and 1 cm² active area. A one-dimensional strip cell architecture eliminates complicated flow behaviors (e.g., potential fluid short circuits, bypass at channel switchbacks, and channel hopping) and minimizes local pressure drop variations in the channel. For these reasons, the strip cell is well-suited for phenomenological, comparative, and detailed model validation studies.

2. Materials and Methods

In this study, a strip cell architecture with only one straight channel and a 1 cm² active area (5 cm long, 0.2 cm wide) was employed, as introduced in previous work [54,55]. Simulations and experiments were conducted on varying channel depth designs (0.25, 0.5, 1, and 2.5 mm) at flow rates of 10–50 mL min^{−1}. A zero-depth channel was not possible for strip cell architecture due to the excessive pressure drop. While enhanced convection is achieved with the shallower channel depths (0.25, 0.5 mm), deeper channel depths (1, 2.5 mm.) were employed to impose a diffusion limited condition in the electrode. Diffusion limitation is conventionally alleviated by increased flow rate; the conditions considered in this work span from 10 to 50 mL min^{−1} cm^{−2}. These area-specific flow rates ranged from conventional to very high. All tests were performed with Nafion® 117 membranes (DuPont™, Wilmington, DE, USA) and carbon paper electrodes (39AA, SGL Group; 280 μm nominal, uncompressed thickness). Flow plates were constructed of BMC 940 (Bulk Molding Compounds, Inc., West Chicago, IL, USA), impermeable to liquids after curing. All channel depth configurations and segmentations were machined in-house.

An electrolyte solution of 1.5 M vanadyl sulfate (Alfa Aesar, 99.9%) in 3.3 M sulfuric acid (Alfa Aesar, ACS grade) was used to perform all tests. Initially, positive and negative side electrolytes (100 mL and 50 mL respectively) were charged at 1.7 V. Cutoff current during charging was 50 mA cm^{−2}. To achieve equal volumes of electrolyte, half of the positive electrolyte was removed after reaching the cutoff current density. Lastly, the electrolyte was galvanostatically discharged to 50% SOC. Electrolyte reservoirs were continuously purged with nitrogen to prevent oxidation of vanadium species (V (II)).

Sensitivity analysis was conducted for various key parameters such as electrolyte density and viscosity, electrode porosity, and permeability. Electrode permeability was found to be the most significant parameter affecting electrolyte velocity distribution in the electrode. However,

since a consistent electrode (carbon paper) and electrolyte composition (1.5 M vanadyl sulfate, 3.3 M sulfuric acid) were employed for all tests, these parameters including permeability were identical for all simulations.

2.1. Polarization Curves and In Situ Localized Current Distribution Measurements

All electrochemical measurements (polarization curves and in situ localized current distribution measurements) were executed over the flow rate range of 10–50 mL min^{−1}; all reported flow rates can be considered area-specific flow rates since the active area was 1 cm². Single-pass polarization curves ensured a constant 50% SOC at the cell inlet. All experiments were conducted potentiostatically by sweeping cell voltage from 1.7 V to 0.2 V in equal increments and recording the current at each step. The maximum current for each cell (at 0.2 V) is considered here as the limiting current. Further details on the hardware, distributed current diagnostic equipment, and other experimental features can be found in previous work [54,61]. The experimental error for current distribution measurements was evaluated via repeated testing and found to be small (maximum ±5%). It was shown in our previous study that lateral current between segments is insignificant [61]. However, it should be noted that the measurement technique is not capable of detecting current spread through the electrolyte due to potential gradients between segments. Details of the technique were provided previously [54].

2.2. Computational Fluid Dynamics

Predictions of the electrolyte flow distribution in the strip cell were obtained using COMSOL Multiphysics® software with a free and porous media flow module [62]. The simulation domain included a channel and porous electrode constructed in three dimensions, as shown in Figure 1. Conservation of mass and momentum were solved to obtain velocity and pressure distributions under the following assumptions: steady state, laminar and incompressible fluid flow, isotropic and homogeneous physical properties. While the fast flow profile in the channel is represented by the Navier–Stokes equations, the Brinkman equations [63] were used to model the comparatively slower flow in porous media (Equations (1) and (2)),

$$\rho(u \cdot \nabla)u = \nabla \cdot [-pI + \mu(\nabla u + (\nabla u)^T)] + F \quad (1)$$

$$\rho \nabla \cdot u = 0$$

$$\rho(u \cdot \nabla)u = \nabla \cdot \left[-pI + \frac{\mu}{\varepsilon_p}(\nabla u + (\nabla u)^T) \right] - \left(\frac{\mu}{\kappa_{br}} + \beta_F |u| \right) u + F \quad (2)$$

$$\rho \nabla \cdot u = Q_m$$

where u is the superficial velocity, p is the pressure, ρ is the density, μ is the dynamic viscosity, ε_p is the porosity of the porous media, κ_{br} is the permeability of the porous media, Q_m is the mass source, F is the body forces acting upon the fluid, and β_F is the Forchheimer drag coefficient. The physical properties for the electrolyte and electrode as well as the other input parameters used for simulation are tabulated in Table 1. The flow field and the electrode dimensions were selected based on the 1 cm² strip cell architecture used in experiments. The electrode porosity and the permeability values for 39AA carbon paper were taken from the manufacturer (SGL) [64]. Considering the compression in the cell, porosity was adjusted to a value of 85% [65]. Electrolyte density and viscosity values were taken from the literature [35,66]. No-slip boundary conditions were assigned for both channel and porous media walls. While the outlet boundary was held 0 kPa, a mass flow rate was specified ($\dot{m}_{in}$) at the inlet. The accuracy of the simulation results is limited by numerical errors due to the discretization of space grids, employed simplifications and assumptions, and imprecision of input parameters. However, the mathematical model predicts experimental data, with a maximum error of 4%.

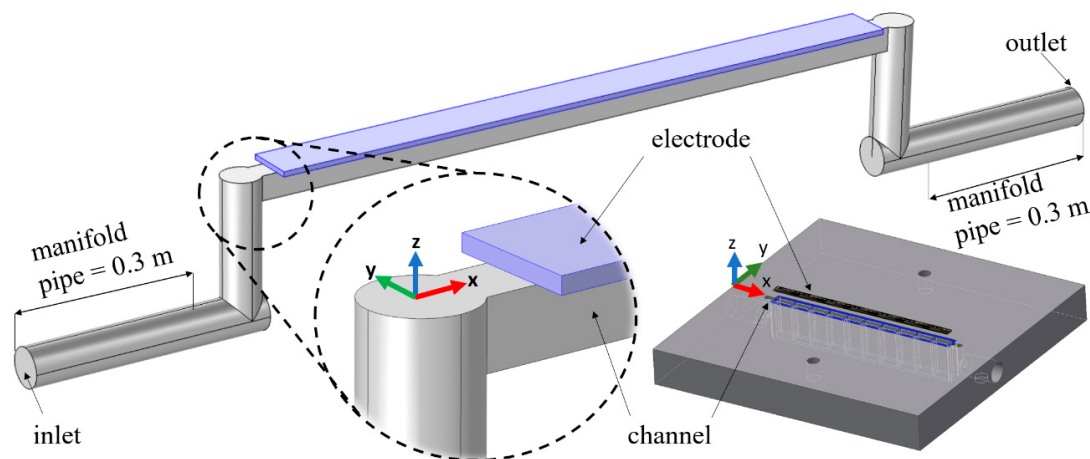


Figure 1. Simulation domains with channel and electrode [56].

Table 1. Simulation parameters.

Parameter	Value	Reference
Channel and electrode length (mm)	50	Chosen
Channel width/land (mm)	1	Chosen
Channel depth (mm)	0.25, 0.50, 1.00, 2.50	Chosen
Electrode width (mm)	2	Chosen
Uncompressed electrode porosity (%)	89	SGL Group [64]
Compressed electrode porosity (%)	85	[65]
Electrode permeability $\times 10^{11}$ (m ²)	1.1	SGL Group [64]
Electrolyte density (kg m ⁻³)	1350	[35,66]
Electrolyte viscosity $\times 1000$ (Pa s)	2.5	[35,66]

3. Results and Discussion

Although CFD with COMSOL utilizes a well-developed, robust numerical simulation of fluid flow, the validity of the results requires experimental validation. Pressure drop measurement is a straightforward verification and has often been used for partial model validation. However, pressure drop discrepancy between experiments and models is a common issue as has been reported in many studies in VRFB literature [22,67]. Kumar et al. attributed this discrepancy to carbon felt electrode intrusion into the channel volume due to compression [67]. However, for the relatively thin carbon paper electrode employed in this work, such intrusion can be considered insignificant. It was observed that non-negligible pressure drop was present in the inlet and outlet tubes located between pressure transducers and the VRFB cell. This pressure drop caused a significant discrepancy between experimental results and the model. To clarify and overcome this issue, inlet and outlet manifold tubes (0.3 m) were included in the simulation domain as seen in Figure 1. The pressure drop due to elbows in the inlet and outlet was also considered even though their contribution was small. Thus, the model accurately represents the entire experimental system. The pressure drop measured across the entire physical domain is defined as overall pressure drop. Computationally predicted and experimentally measured overall pressure drops [54,55] were compared for all strip cell configurations (0.25, 0.50, 1.00, and 2.50 mm) and a range of flow rates from 10 mL min⁻¹ up to 50 mL min⁻¹, as shown in Figure 2. Good agreement was achieved between the numerical predictions and the experimental measurements with a maximum error of 4%.

It has been reported in previous work that enhanced electrochemical performance and relatively uniform current density distribution can be attributed to increased convective flow in the porous electrode [54]. Figure 3a–d show predicted electrolyte velocities at the midline of a channel and adjacent electrode for all strip cell configurations (0.25, 0.50, 1.00, and 2.50 mm depth) and flow rates (10, 20, 30, 40, 50 mL min⁻¹). Since velocity in the electrode was much smaller than in the channel,

a magnified view of the fluid velocity in the porous layer is included in Figure 4a–d. It is clear that electrolyte velocities both in the channel and electrode change as a function of channel depth and flow rate; more shallow cell configurations at elevated flow rates have the highest average velocities. Computationally predicted average electrolyte velocities both in channel and electrode are tabulated in Table 2. These velocities were obtained by averaging the surface on the midplane of a channel and adjacent electrode.

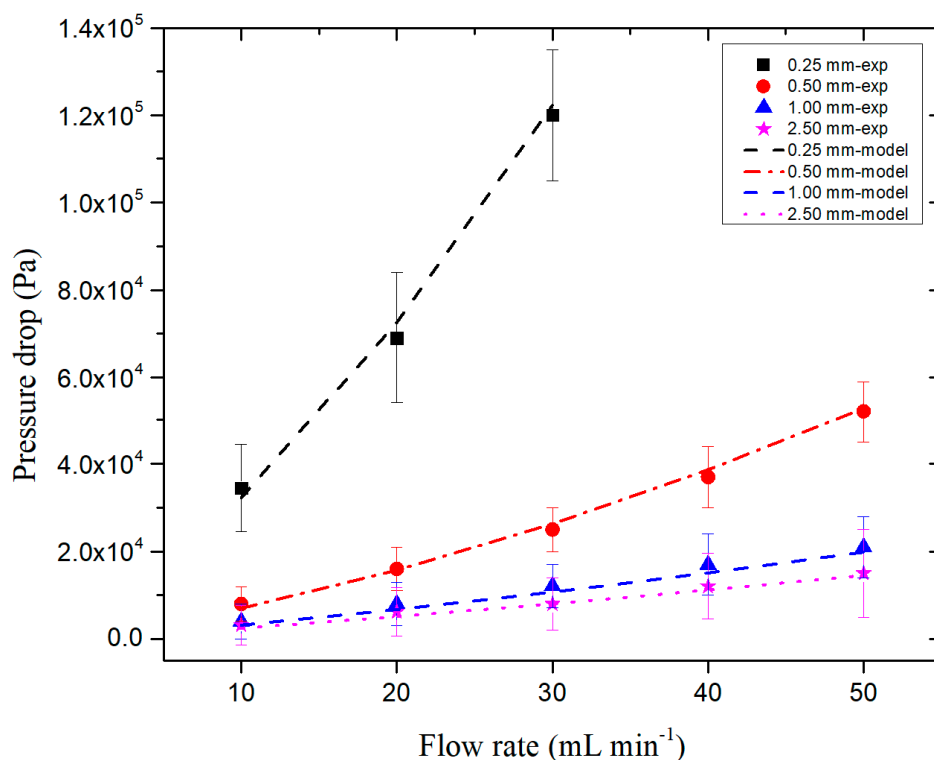


Figure 2. Computationally predicted and experimentally measured pressure drops through the strip cell, including inlet and outlet manifold pipes. Error bars reflect both pulsing from the peristaltic pump and experimental variability.

In addition to electrolyte flow results, Table 2 also shows experimentally measured maximum current densities at 0.2 V hold. While the electrochemical performance of the cell was directly proportional to the electrolyte velocity in the electrode, there was no detectable, direct correlation between electrolyte velocity in the channel and electrochemical performance. Figures 3e and 4e show the average channel velocity/maximum current density and average electrode velocity/maximum current density correlations, respectively. Even though the electrolyte velocities in the electrode were much smaller than the velocities in the channel, a small enhancement of velocity in the electrode yielded significant electrochemical performance improvement. According to Faraday's second law of consumption and production of species, the quantity of reactant consumed is directly proportional to the charged passed [65]. In VRFB literature, volumetric electrolyte penetration into the electrode is usually assumed as an amount of reactant consumed under limiting conditions [37,40,59]. Thus, greater electrolyte penetration into the electrode corresponds to greater electrolyte velocity in the electrode; higher current is generated in the cell as a result. This effect occurs because increased velocity in the electrode reduces the diffusion boundary layer thickness around the fiber surfaces, enhancing maximum transport rates to the reaction surface [45]. Rapidly replenished fresh electrolyte also lowers concentration polarization in the electrode. This allows cells to operate at a higher current density with more uniform current distribution. However, mass transport in the electrode is limited by the diffusion resistance between the liquid electrolyte and the fiber surfaces at the pore level, even at very high electrolyte velocity in the electrode; this resistance can be mitigated but never completely removed.

Figure 4e illustrates that, as electrolyte velocity in the electrode increases, the relative increase in current density attenuates, and the rate of increase in limiting current gradually flattens out. Additional data were not possible for higher velocities or shallower channels due to excessive pressure drop; but it is clear that maximum limiting current is a direct function of electrolyte velocity in the electrode (e.g., reactant convection). Up to the maximum current, however, increased electrolyte velocity in the electrode improves the electrochemical performance; the current distribution also becomes more uniform, an indicator of sufficient mass transport to the electrode surfaces.

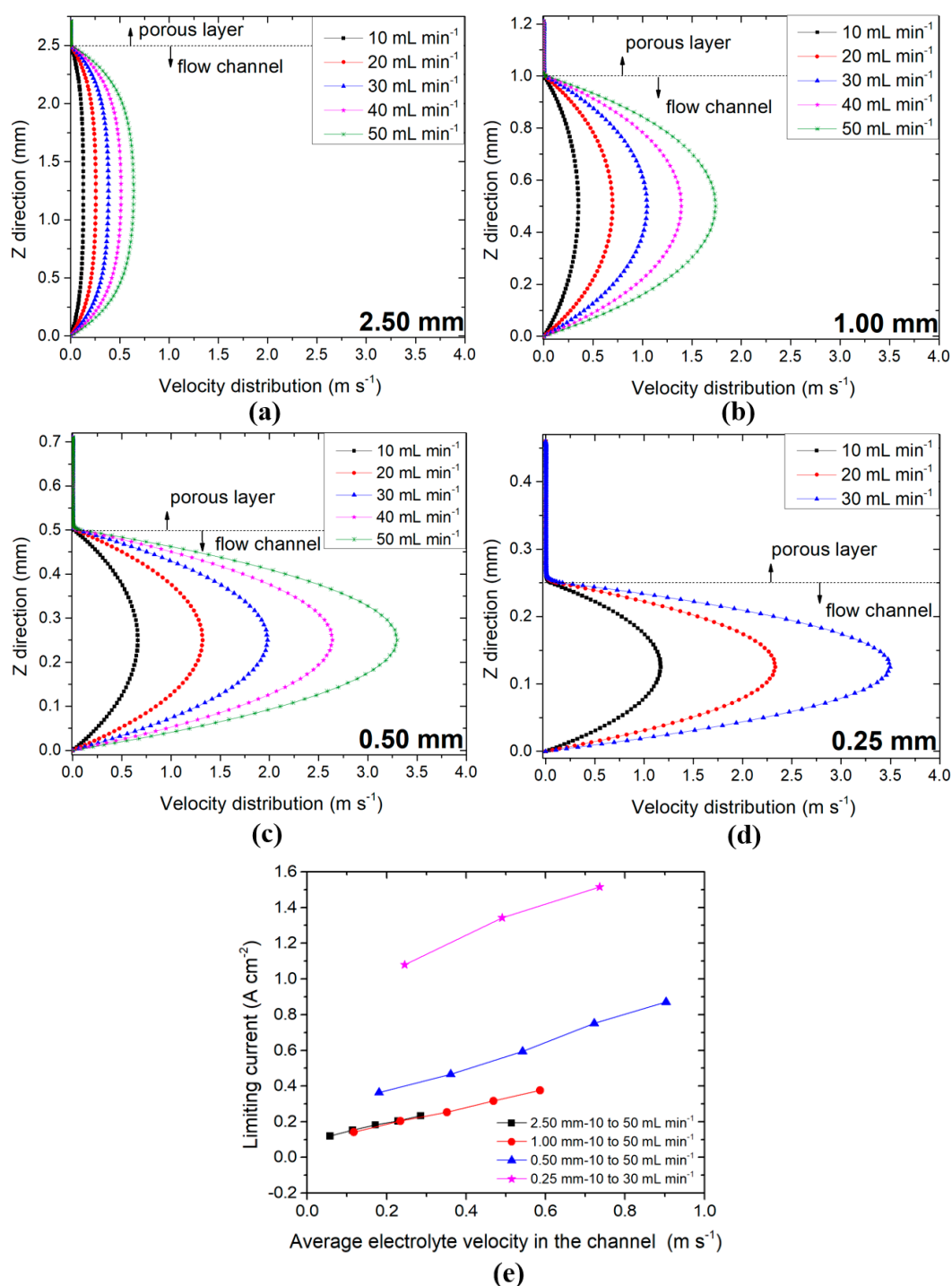


Figure 3. Predicted electrolyte velocity distribution at a cut line in the center of the electrode and channel domain: (a) 2.5 mm (b) 1 mm (c) 0.50 mm (d) 0.25 mm and (e) correlation between average channel velocity and measured limiting currents.

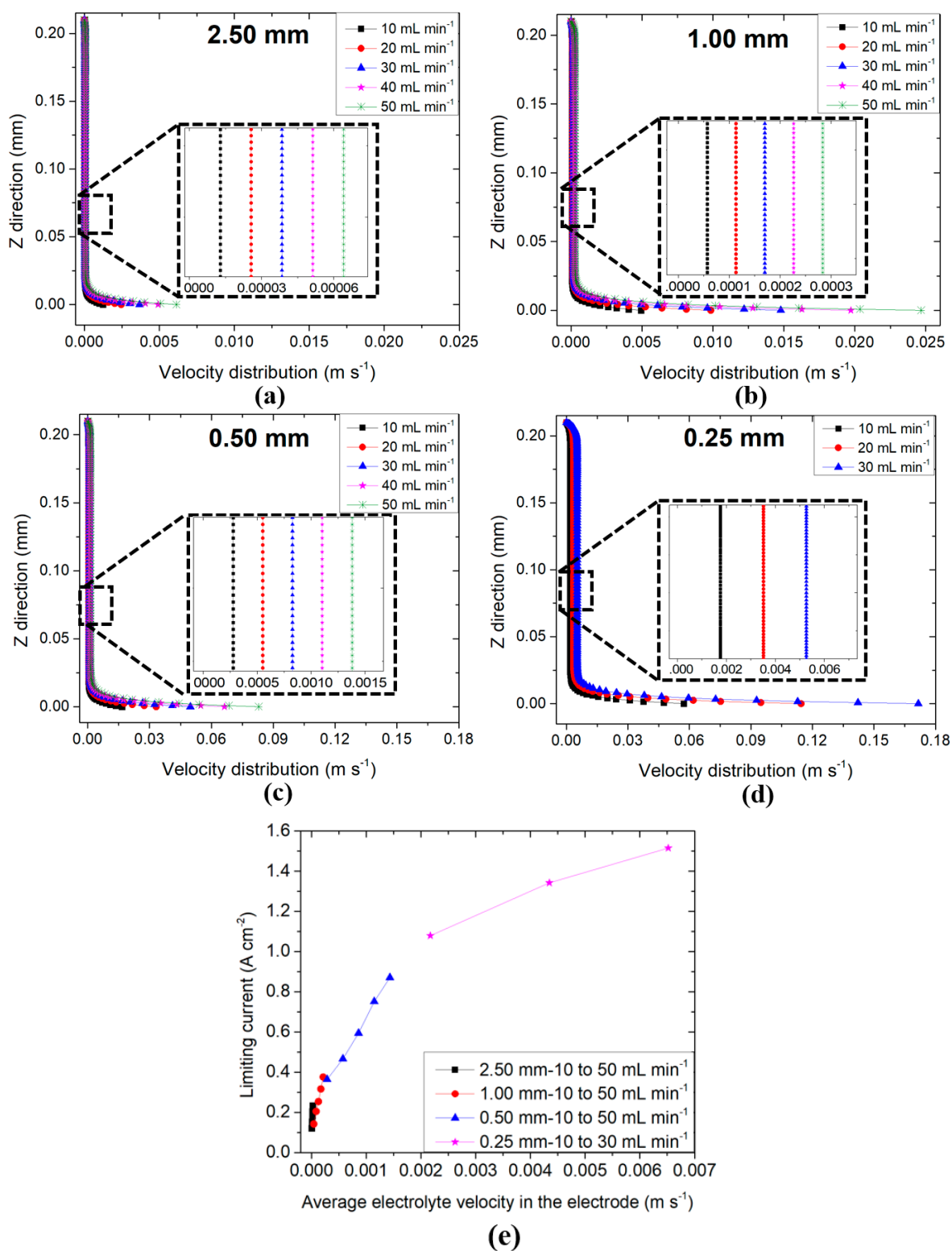


Figure 4. Magnified view of predicted electrolyte velocity distribution in the electrode: (a) 2.50 mm (b) 1.00 mm (c) 0.50 mm (d) 0.25 mm and (e) correlation between average electrode velocity and measured limiting currents.

Table 2. Selected parameters for the range of channel depths and flow rates considered in this work. Limiting current is the current achieved at 0.2 V discharge for a 50% SOC electrolyte.

Channel Depth (mm)	Flow Rate (mL min ⁻¹)	Average Velocity in the Channel (m s ⁻¹)	Average Velocity in the Electrode (m s ⁻¹)	Limiting Current (A cm ⁻²)	Average Local Pressure Drop (Pa)	Overall Pressure Drop (Pa)
2.5	10	0.057013	4.85×10^{-6}	0.1291	18.8	2433
	20	0.11402	9.71×10^{-6}	0.1528	37.7	5114
	30	0.17104	1.45×10^{-5}	0.1825	56.6	8029
	40	0.22805	1.94×10^{-5}	0.2044	75.5	11,167
	50	0.28505	2.42×10^{-5}	0.2337	94.4	14,510
1.00	10	0.11723	4.27×10^{-5}	0.1419	82.8	3157
	20	0.23446	8.55×10^{-4}	0.2044	165.6	6726
	30	0.35169	1.28×10^{-4}	0.2633	248.3	10,693
	40	0.46892	1.71×10^{-4}	0.3161	331.1	15,065
	50	0.58612	2.14×10^{-4}	0.3754	414.1	19,838
0.50	10	0.18066	2.86×10^{-4}	0.3638	402.3	6992
	20	0.36131	5.73×10^{-4}	0.5424	803.8	15,725
	30	0.54197	8.60×10^{-4}	0.5939	1205.5	26,307
	40	0.72263	1.15×10^{-3}	0.7513	1607.4	38,666
	50	0.90325	1.43×10^{-3}	0.8704	2010.2	52,679
0.25	10	0.24549	2.17×10^{-3}	1.0791	2537.1	32,264
	20	0.49097	4.35×10^{-3}	1.342	5071.1	72,569
	30	0.73646	6.52×10^{-3}	1.5150	6340.3	122,290

Figure 5 shows the impact of electrolyte velocity on the current distribution at 50% SOC for selected flow rate/channel depth configurations. These configurations were chosen among eighteen different flow rate/channel depth combinations to demonstrate current distribution variations more clearly. Figure 5a shows absolute local current distribution as a scatter plot while the contour plots in Figure 5b show percent deviation from the average current. Although the impact of increased electrolyte velocity on the measured local current is apparent in Figure 5a, relative differences in current down the channel are less clear due to the different current scales. Thus, the current measured in each segment was normalized to the average current for any configuration to quantify current distribution down the channel. It is clearly seen in Figure 5b that the current distribution down the channel became relatively uniform as electrolyte velocity in the electrode increased. While the lowest predicted velocity in the electrode (2.5 mm channel depth at 10 mL min⁻¹) had the largest current deviation (+102% to −33%), the highest predicted velocity in the electrode (0.25 mm channel depth at 30 mL min⁻¹) had the lowest current deviation (+2% to −8%) from inlet to outlet. Nonuniform current distribution has been reported in recent studies in VRFB literature. Houser et al. attributed different current distribution patterns for serpentine and interdigitated flow field designs to disparate velocity gradients in the electrode [22]. However, variations in the electrolyte velocity in the electrode for a strip cell are negligibly small; thus, the velocity gradients in Houser et al. [21] can be attributed to higher-order behaviors not present in the 1-D strip cell. Figure 6 shows predicted velocity distribution in the electrode for all channel depths (0.25, 0.50, 1.00, and 2.50 mm) at a 30 mL min⁻¹ flow rate. Velocity distributions down the channel for all channel depths were highly uniform. Thus, the large current gradients for experiments with low fluid velocity are attributed to local concentration depletion in the electrode; such concentration depletion leads to diffusion limitation. A natural next step, then, is calculation of active species concentration down the channel in the electrode region.

It has been shown that the electrochemical performance and current distribution can be correlated to the hydrodynamics in the electrode. Development of a correlation between pressure drop and electrochemical performance is thus of interest, since overall pressure drop is the primary drawback

to increased flow rates, which are known to enhance performance. Achieving enhanced velocity in the electrode (and thus mass transport via convection) with minimal increases in overall cell pressure drop is a potential avenue to greater overall efficiency. Understanding this correlation is essential to maximize net system efficiency due to the existence of parasitic pumping losses. Table 2 shows both experimentally measured maximum current densities and computationally predicted overall pressure drops for all channel depths and flow rates in this study. It can be noted that two configurations, 0.5 mm-10 mL min⁻¹ and 1.0 mm-50 mL min⁻¹, yielded very similar electrochemical performance. While the channel depths, flow rates, and predicted overall pressure drops (6992 Pa and 17,917 Pa) were quite different, these two configurations yielded very similar electrode fluid velocity (1.26×10^{-4} m s⁻¹ and 1.71×10^{-4} m s⁻¹) and, as hypothesized, very similar current density (0.3638 and 0.3754 A cm⁻²) at the given condition. This result indicates that, to some degree, performance can be enhanced while suffering reduced pressure drop penalty using optimized architecture and operating parameters.

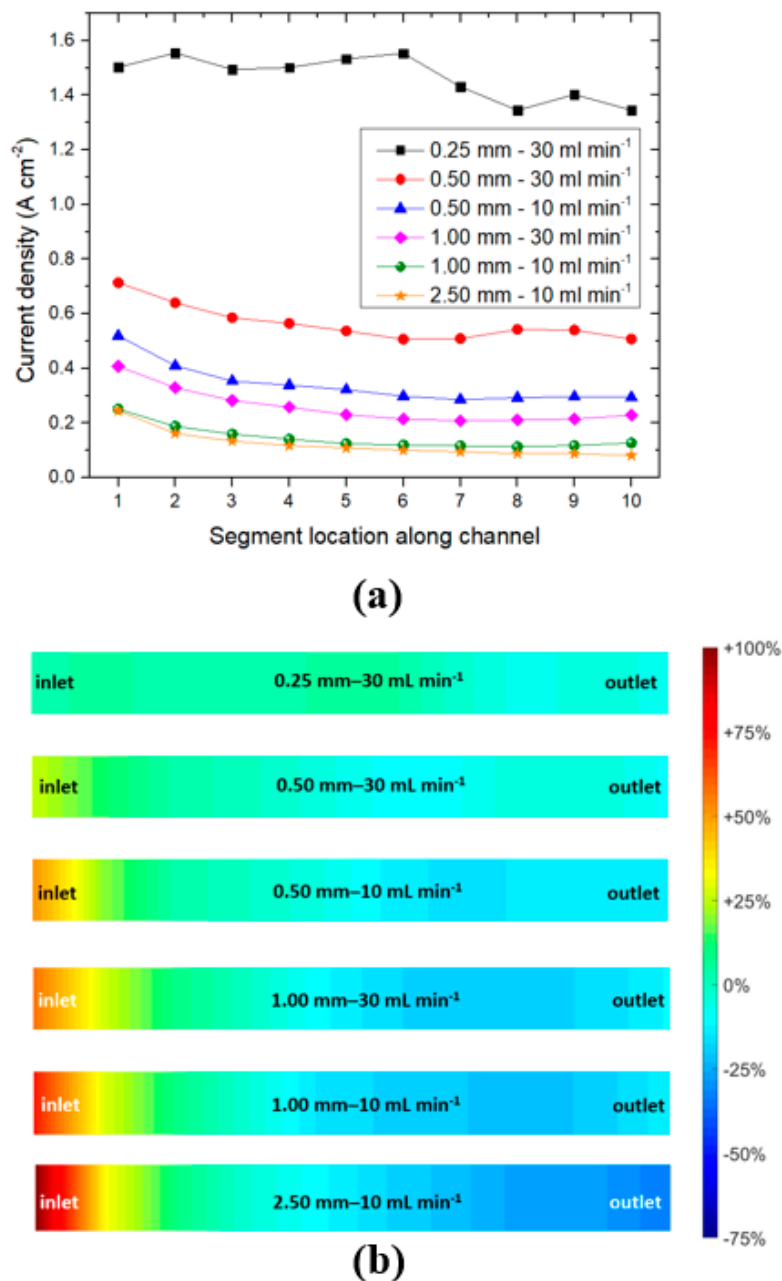


Figure 5. Measured local current (a) and (b) normalized current distribution comparisons at 50% SOC, 30 mL/min, 0.2 V hold.

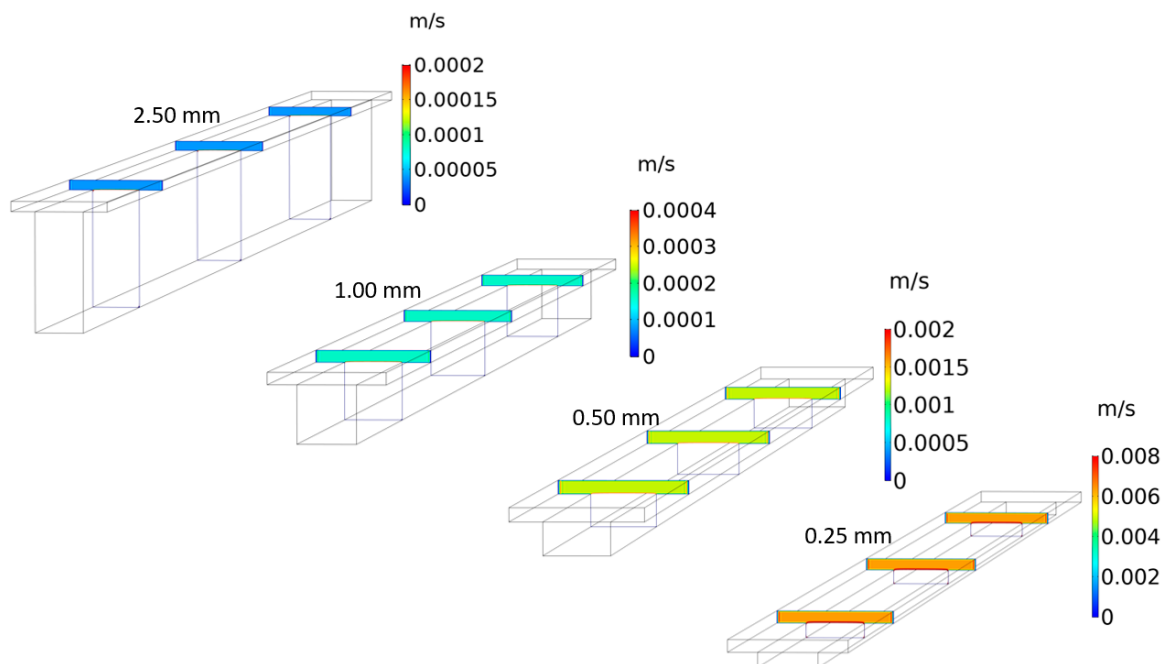


Figure 6. Predicted electrolyte velocity distribution at a cut plane through the electrode domain for all channel depths at a 30 mL min^{-1} flow rate.

To investigate this pressure drop discrepancy and understand the relationship between pressure drop and electrochemical performance, computationally predicted pressure distributions for these configurations are compared in Figure 7. Figure 7a shows pressure distribution from inlet to outlet, including elbows and plumbing tubes. While the color code stands for the pressure gradient from inlet to outlet, the arrows indicate the local pressure drop defined as the pressure drop through each individual segment down the channel. The pressure gradient for the configuration with $0.5 \text{ mm-}10 \text{ mL min}^{-1}$ appears qualitatively steeper than the pressure gradient for $1.0 \text{ mm-}50 \text{ mL min}^{-1}$ at the channel/electrode region. However, it should be noted that these configurations have different pressure scales. Local pressure drops indicate that the pressure gradient is very similar down the channel for these configurations. The average local pressure drop is predicted to be 436 Pa and 525 Pa for the configurations with $0.5 \text{ mm-}10 \text{ mL min}^{-1}$ and $1.0 \text{ mm-}50 \text{ mL min}^{-1}$, respectively. For another perspective on this behavior, CFD simulations were performed excluding the elbows and plumbing tubes, effectively capturing only the channel electrode region, called “simplified geometry” here. Figure 7b shows that the pressure gradient from channel inlet to outlet is identical for both configurations. Although the average local pressure drop for the simplified geometries is underpredicted (402 Pa and 414 Pa), it indicates that the local pressure drop in the complete system is primarily caused by a combination of flow rate and channel depth, as would be expected. The overall pressure drop discrepancy between the two configurations is thus attributed to pumping different flow rates (10 mL min^{-1} and 50 mL min^{-1}) through the same manifolding before and after the active area. The local pressure drops (for simplified geometries) for all flow rates and channel depths are tabulated in Table 2. It is found that the local pressure drop is also directly proportional to the electrode velocity as seen in Figure 8a. This correlation indicates that the local pressure drop is the driving force for electrolyte to penetrate into the electrode region. It is also seen in Figure 8a that shallower channels instigate electrolyte penetration more effectively than deeper channels. Considering a fixed local pressure drop (e.g., $3 \times 10^3 \text{ Pa}$) for all channel depths, it is possible to achieve three times faster electrode velocity by employing the shallowest channel configuration. Conversely, there is no direct relationship evident between local pressure drop and average channel velocity as seen in Figure 8b. Figure 8c,d show local pressure drop–current and overall pressure drop–current correlations, respectively. As expected, the local pressure drop–current correlation in Figure 8c is very similar to the

average velocity–current correlation in Figure 4e. Figure 8d indicates that the pressure drop arising due to any fluid path outside the channel electrode region (e.g., external plumbing) does not contribute to the current and must be minimized to improve overall system efficiency of the VRFB cell.

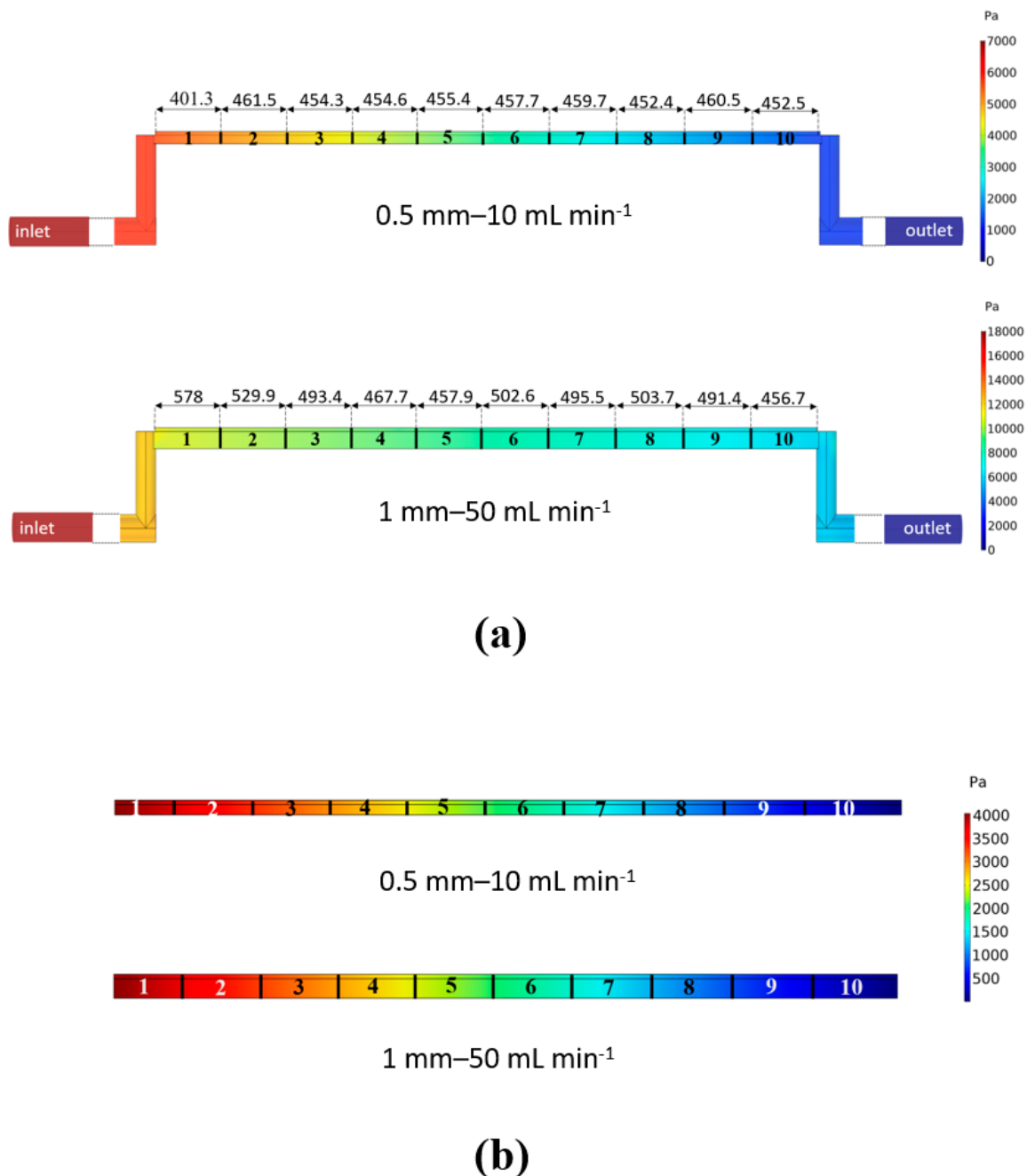


Figure 7. Predicted pressure drop distribution for (a) complex and (b) simplified geometries.

It can be concluded that improved VRFB electrochemical performance can be achieved by optimizing the tradeoff between pressure drop and in-electrode velocity of the electrolyte. Deeper channels are found to be less effective at increasing convection in the electrode, even at higher flow rates in the range studied here (up to 50 mL/min/cm²). Additionally, the overall pressure drop needed to achieve high electrolyte penetration in the electrode is much greater for deeper channels than shallower channels. Considering the manifolding needed for VRFB stack application, operating at lower flow rates can significantly decrease parasitic pumping losses. It is also observed that the pressure drop suffered in the channel is effectively wasted because electrochemical performance is

shown to not correlate to the channel velocity. Thus, flow fields and electrodes should be designed to enable maximum electrolyte velocity in the electrode with minimal overall pressure drop; shallow channels are one avenue to this goal.

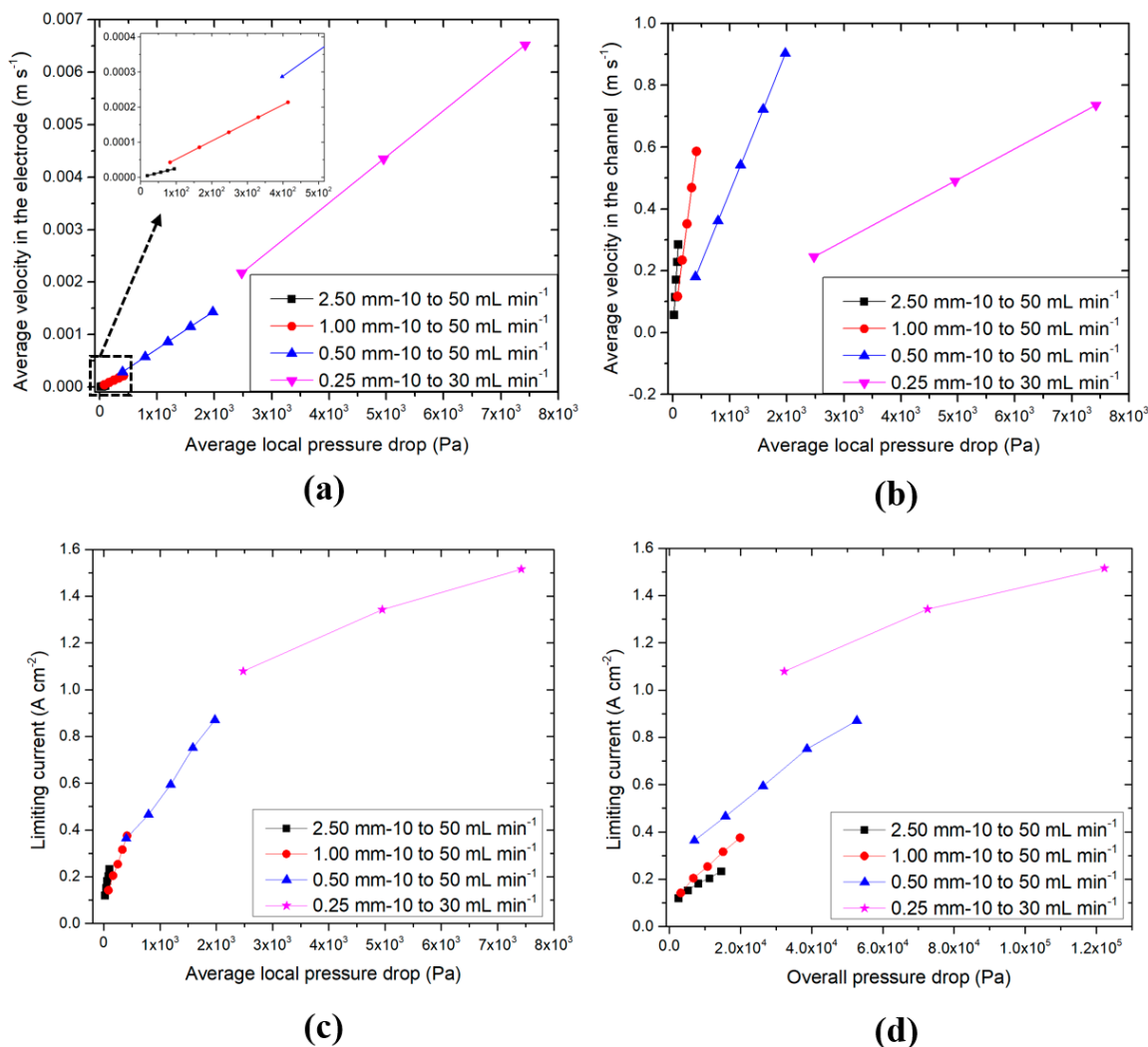


Figure 8. Correlations between (a) local pressure drop and average channel velocity, (b) local pressure drop and average electrode velocity, (c) average local pressure drop and limiting current, and (d) overall pressure drop and limiting current.

4. Conclusions

Convection in the electrode is a critical mechanism for transport of active species to/from the reaction surfaces in a high-performance VRFB. Thus, the impact of convection on electrochemical performance and pressure drop must be understood to achieve enhanced electrochemical performance while minimizing pumping losses. The simple 1-D strip cell design employed for this study minimizes local pressure drop variations and achieves relatively uniform electrolyte velocity distribution down the channel, and is thus configured to provide precise benchmark data. A range of electrolyte velocities inside the cell was achieved by employing various channel depths (0.25, 0.5, 1, 2.5 mm) and flow rates (10–50 mL min⁻¹). Velocity and pressure drop distributions down the channel were predicted via CFD simulation, while localized current distribution was measured. It was found that the experimentally measured maximum current for each configuration scaled linearly with the predicted electrode region average fluid velocity. However, there was no direct correlation observed between electrolyte velocity in the channel and the limiting current. Increasing local electrolyte velocity in the

electrode facilitates active species transport, reducing the diffusion boundary layer thickness around the fiber surfaces. This insight guides design for enhancing flow into the electrodes. Employing shallow channels is a practical way to improve convective flow in the electrode while suffering a comparatively moderate pressure drop penalty, with limitations in the extreme. It is demonstrated that there is a point where electrochemical current cannot be improved with further flow velocity increases, which is likely due to the existence of diffusion resistance between the liquid electrolyte and the fiber surfaces at the pore level. Current distribution tests indicate that increased electrolyte velocity limits local mass transport limitations in the electrode, resulting in more current uniformity down the channel. Although the electrolyte velocity distribution in the electrode is homogeneous, high current deviations are observed from inlet to outlet for experiments with low fluid velocity. Local concentration depletion in the electrode is speculated to be responsible for these large current gradients. As a result of this work, prediction of electrolyte velocity in the electrode can be correlated with experimentally measured current distribution; modeling work can thus focus on linking local properties in the electrode (e.g., concentration and fluid velocity) to electrochemical performance.

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