Membraneless Micro Redox Flow Battery: From Vanadium to Alkaline Quinone

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This work presents the first proof-of-concept of a membraneless micro redox flow battery with an automated closed-loop control. Using micro actuators and micro sensors, charge and discharge is achieved in continuous operation in recirculation. A maximum value of 60% State of Charge with commercial Vanadium electrolyte, monitored via online spectrometry, is delivered, with a maximum discharge current density of 60 mA/cm², and a discharge volumetric capacity of 21.5 Ah/L. Next, cycling tests show a decrease in coulombic efficiency of 0.5% per cycle, and capacity loss. To overcome some drawbacks encountered with Vanadium, an alkaline quinone electrolyte is used instead in the membraneless micro redox flow battery.

Using this new electrolyte, sixty continuous cycles are performed showing a decrease in coulombic efficiency of 0.6% per cycle, and a capacity loss of only 1.2% per cycle. The performances obtained outshine previous literature results. The highest energy efficiency ever obtained for a membraneless micro redox flow battery is presented here with alkaline quinone having an efficiency of 28.9%. The cycling of a membraneless micro redox flow battery is successfully performed for the first time. This work also includes performance improvement suggestions for future work, with this landmark opening a promising path towards achieving the metrics of conventional redox flow batteries.

Introduction

Microfluidic technology has expanded significantly in recent years, not only to conventional Lab-on-a-Chip applications, but to a greater variety of energy storage applications, such as fuel cells, flow batteries, electrolysis cells, hybrid cells and other emerging devices. This growth demonstrates the potential use of this technology for enhanced electrochemical energy storage as a crucial complement for renewable energy transition.[1–4]

New concepts of microfluidics in the development of redox flow batteries entail the most disruptive advance for this technology during the last years.[5–8] The presence of a

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membrane in conventional redox flow batteries presents drawbacks, such as costs increase from the economical point of view, and a decrease in battery performance due to the addition of extra internal resistance on the cell.[9] Microfluidic technology allows the removal of this membrane by using laminar flows. Membraneless electrochemical systems have significant advantages, since membranes account for 22% to 40% of the stack cost, lifetime limitations, and maintenance costs.[10–12] Moreover, the application of microfluidics concepts allows to build more compact global modules that can be stacked to easily scale-up the technology.[13] This would, improve performance, while avoiding shunt currents, provide a more homogeneous use of electrodes, and increase chemical gradients for reactions, among other benefits.[1]

In addition, the most employed chemistry for commercial redox flow batteries is the all-vanadium redox flow battery, utilizing vanadium-based electrolytes in strong acidic solutions.[14] Despite its competitive electrochemical performance for stationary applications, concerns arise regarding vanadium supply chain control,[15] and its corrosive nature. This has prompted the exploration of alternative chemistries with more accessible raw materials based on redox active organic molecules that are non-toxic, non-flammable, and safe.[16,17]

The combination of vanadium chemistry and microfluidics for energy conversion has seen significant advances in recent years. Researchers have dedicated efforts towards developing various energy conversion systems since the early 2000s.[5] Concurrently, research groups have explored this type of microfluidic systems, substituting vanadium chemistry with alternative organic compounds.[8,18,19] These works have involved extensive optimization of reactor designs with the aim of enhancing system performance.[4,20]

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Most studies have focused on utilizing vanadium as the charged fuel passing through microfluidic reactors to obtain energy operating as a fuel cell, with only one exception developed by Lee et al.,[7] which employs a two-step syringe pumping system for charge and discharge in one single pass for each one. All these efforts towards energy conversion using microfluidics, had not resulted yet in a rechargeable battery, limited usually by the low performance of the system used.

Moreover, despite significant progress, none of the previous works have employed a closed-loop fluidic system with electrolyte recirculation like the ones used in conventional flow batteries, except for the recent work of Oraá-Poblete et al.[21] which was one of the first steps toward rechargeability of a membraneless micro redox flow battery.

Thanks to all these efforts made in the electrochemistry and microfluidics fields, this study presents for the first time a rechargeable membraneless micro redox flow battery operating in recirculation mode. This innovative approach not only enables charge-discharge processes using vanadium electrolyte but also demonstrates the recirculation of the electrolyte and cycling operation. Furthermore, as mentioned earlier, some of the limitations of vanadium electrolyte are mitigated by exploring alternative compounds. Hence, the initial steps towards incorporating organic chemistry into redox flow micro batteries by using an alkaline quinone-based electrolyte are introduced.

Results and Discussion

Cell Characterization

The membraneless Micro Redox Flow Battery used in this research is based on the one presented by Oraá-Poblete et al.[21] with an improvement of the electrical external contacts. The details of reactor design and microfluidic system are explained in S1 of Supporting Information. For the electrochemical characterization, commercial Vanadium electrolyte supplied by Oxkem Limited (1.8 M concentrated) was used, and polarization curves were obtained by varying the flow rate of both electrolyte streams (negative and positive). Polarization curves shown in Figure 1 offer a comprehensive insight into the electrochemical behavior of the reactor, showing the obtained current density against its corresponding power density, where the potential is increased from 0 to 1.5 V by steps of 0.1 V. Within this electrochemical framework, a peak power density output of 155 mW/cm² corresponding to 208 mA/cm² for the higher flow rate applied of 300 µL/min is obtained.

Results are compared with those presented by Kjeang et al.[6] operating in fuel cell mode, which is the first design using porous electrodes architecture, those obtained by Lee et al.[7] operating in one single pass charge-discharge, and with results presented by Ibrahim et al.[22] which were the highest performance reported up to date for a Vanadium micro reactor without improving electrolyte conductivity.[23] Improvement of the micro reactor performance presented here is achieved only through better external electrical contact, and a precise fluid
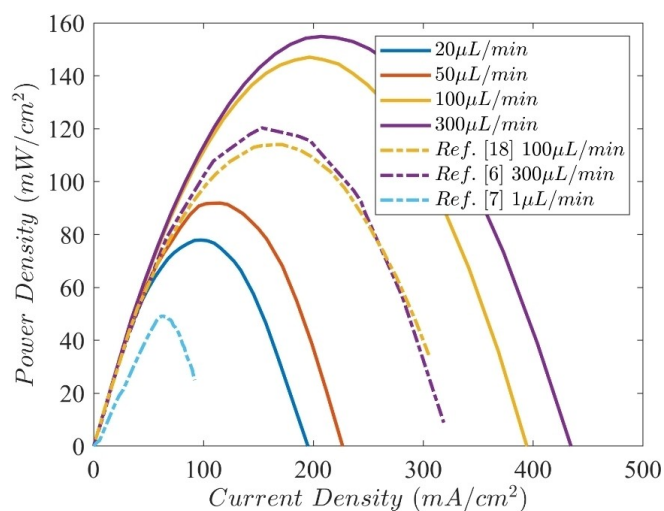
Figure 1. Polarization curves of membraneless microfluidic vanadium redox flow battery for flow rates 20, 50, 100, 300 µL/min.
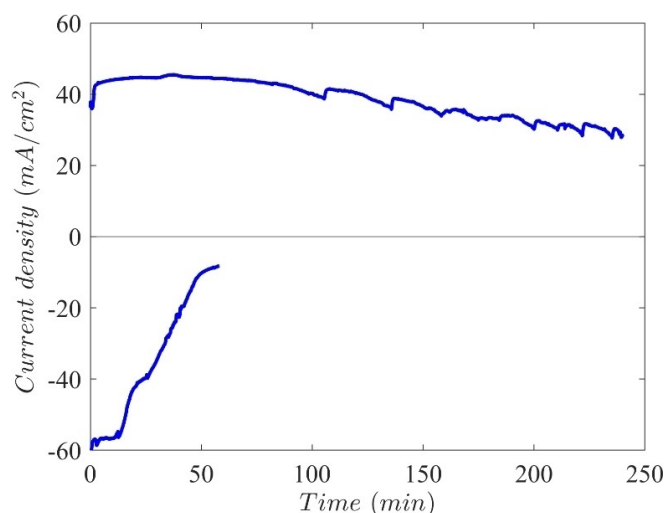
control due to an integrated flow control system which has been proven critical for the performance of membraneless micro redox flow batteries.[24]

Charge-Discharge of Membraneless Vanadium Micro Redox Flow Battery (MVMRFB)

A total volume of 400 µl of Vanadium electrolyte was fed in each stream (positive and negative), flowing directly V^{3+} at the negative side and V^{4+} at the positive side (0% State of Charge). Flowing in a closed loop, a charge-discharge experiment was carried out in inert atmosphere (Argon), to avoid V^{2+} self-discharge in contact with oxygen. The charge process operated at constant 1.7 V in a potentiostatic mode for 4 hours. Charge operation was run at high voltage to reduce operation times, and to obtain the fastest conversion of species in the electrodes to compensate self-discharge losses by specie crossover in the miscible interface due to absence of a physical barrier and direct contact of anolyte and catholyte inside the micro reactor. This allows to obtain the maximum possible value for State of Charge until the self-discharge compensates the charge.

Next, the discharge was performed by applying 0.8 V in potentiostatic mode for about 1 hour. Charge-discharge operation was carried out with constant flow rates of 40 µl/min for anolyte and 15 µl/min for catholyte thanks to the implementation of an automated closed-loop control. Flow rates were selected based on previous research indicating that reactions taking place in the negative side are responsible of poor performance due to mass transport limitations and side reactions during V^{3+} reduction process,[25,26] as well as following results previously obtained for preconditioning process in membraneless Vanadium micro redox flow batteries.[21]

Figure 2 shows the current density obtained during charging at 1.7 V, the current decreases with time, as charging progresses. In addition, current density for the corresponding discharge at constant 0.8 V of the battery shows decreasing

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Figure 2. Current profile obtained for the potentiostatic charge (1.7 V) and discharge (0.8 V) of the MVMRFB. (Current fluctuations during charging may result from viscosity changes or bubble formation)

current with time, as the discharge process occurs because V^{2+} and V^{5+} species generated during charge are being consumed. From Figure 2, the charge and discharge capacity evolution could be calculated, giving a 39.4 mAh for the charge and 8.6 mAh for the discharge, which corresponds to an achieved coulombic efficiency of 21 %. Considering the capacity obtained during discharge and given the Vanadium concentration, it yields an electrolyte utilization of 43 %.

The State of Charge evolution of both anolyte and catholyte was monitored in real time independently during the whole process by UV-visible spectroscopy as mentioned in S1. It followed the calibration described by Maurice et al.[27] and is presented in Figure 3 for both negative and positive streams. At the beginning of the charge process, negative State of Charge, was lower than positive one. This happens for the first 50
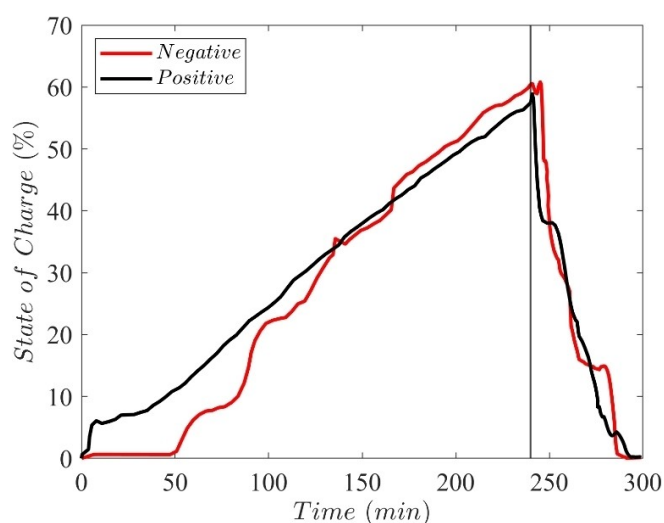
Figure 3. State of charge evolution for negative and positive MVMRFB streams for the charge-discharge process as measured via UV-vis spectroscopy.

minutes of operation, and it could be related to a slower conversion from V^{3+} to V^{2+} than from V^{4+} to V^{5+} which could imply the presence of side reactions such as hydrogen evolution. However, it could also be caused by a non-homogeneous mixing of species in the electrolyte stream, due to internal dead volumes in the fluidic loop as well as flow stratifications, these could delay State of Charge readings.

A maximum State of Charge of around 60 % is achieved in the negative and positive electrolytes after 4 hours of charge. At this point bubbles appeared in the system, likely arising from hydrogen evolution being favored by the decrease of V^{3+} concentration available in the negative electrode, therefore, discharge at constant 0.8 V was followed.
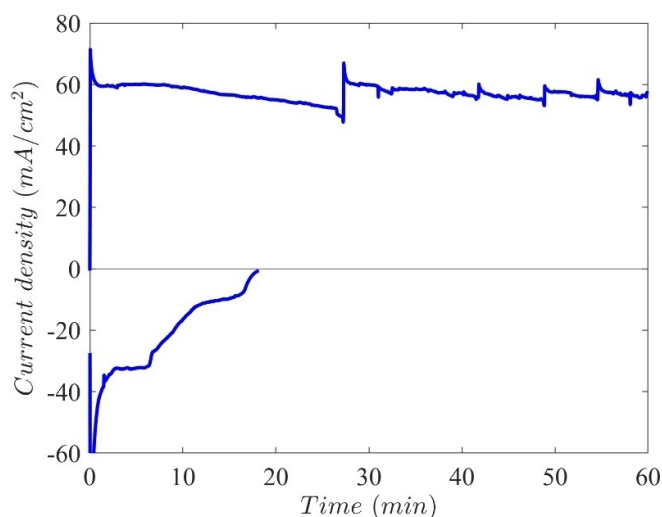
Cyclability of Membraneless Vanadium Micro Redox Flow Battery

Despite previous reported results, a redox flow battery using a microfluidic system in closed fluidic loop has never been cycled before. The closest attempt was a single charge and discharge in one pass through the micro reactor presented by Lee et al.[7] with a system that does not allow continuous operation as in conventional redox flow batteries. In this section, we present for the first time a MVMRFB cycling with a microfluidic system. For this test, while the electrolyte is recirculating in the system at 100 μl/min for catholyte and 30 μl/min for anolyte, a charge potential of 1.7 V during 1 h was applied, and after that, a discharge potential of 0.8 V was applied until the current obtained is zero.

Real time measurements of State of Charge with spectroscopy are not included in this experimental setup for simplicity and also due to limitations discussed in the previous section regarding delayed and false readings due to non-homogeneous mixing that must be overcome. It is worth mentioning that mixing problems of the streams must be tackled in near future operations, the aim is to integrate micro-optical sensors as part of the battery management system to improve performance of the device.

In this experiment, the depth of charge is lower to avoid side reactions and a high self-discharge. For this reason, the discharge is considered finished when the current is zero, since it is assumed that there is no imbalance between the positive and negative States of Charge due to the absence of side reactions and the total control of self-discharge by the integrated flow control system. Bubbles were not observed after 14 hours of operation of the MVMRFB, which indicates the absence of significant hydrogen evolution. Figure 4 shows the current profile of the first cycle.

The obtained current density (57 mA/cm^2 for charge) is higher than the one from the previous section due to an improved contact resistance of the micro reactor. In addition, a lower charge operation time was selected, so the electrolyte utilization will be lower (8.64 % for this cycle). A high Vanadium concentration (as the 1.8 M in commercial electrolyte) implies longer operation times to reach a given State of Charge with the micro reactor used here. Longer operation times yield

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Figure 4. Current profile obtained for the first cycle of the MVMRFB. 1.7 V and 0.8 V were applied for charge and discharge processes, respectively.

higher self-discharge by diffusion of species at the interface driven by a higher gradient of concentrations between both streams (positive and negative). This led to lower coulombic efficiencies, such as the one obtained in the experiment described in the previous section (20%). For the first cycle presented in this section it is 11.67%. Coulombic efficiencies in membraneless micro redox flow batteries for highly concentrated electrolytes must be improved. After the first cycle, the MVMRFB operated ten more cycles with the performance shown in Figure 5.

The initial cycle exhibits the highest capacity (4.17 Ah/L), coulombic efficiency (11.67%) and discharge power (17.58 mW/cm^2). During the successive ten cycles, a gradual decrease in capacity is observed, averaging 7.76% capacity loss per cycle from the initial recorded value. Remarkably, the decrease in coulombic efficiency per cycle is 0.48%. Detailed insights into current, power, energy, electrolyte utilization, and efficiencies
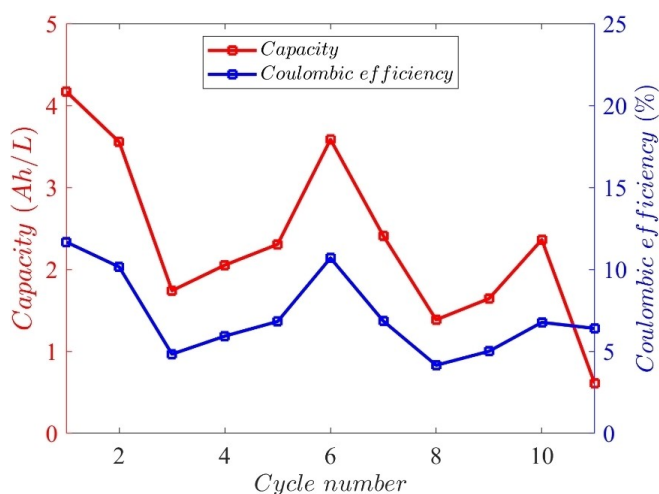
Figure 5. Cyclability results of the MVMRFB with commercial Vanadium electrolyte (1.8 M).

(both energy and coulombic) across six significant cycles (1st, 3rd, 7th, 9th, 11th) are available in Table S1 of Section S2 in the Supporting Information.
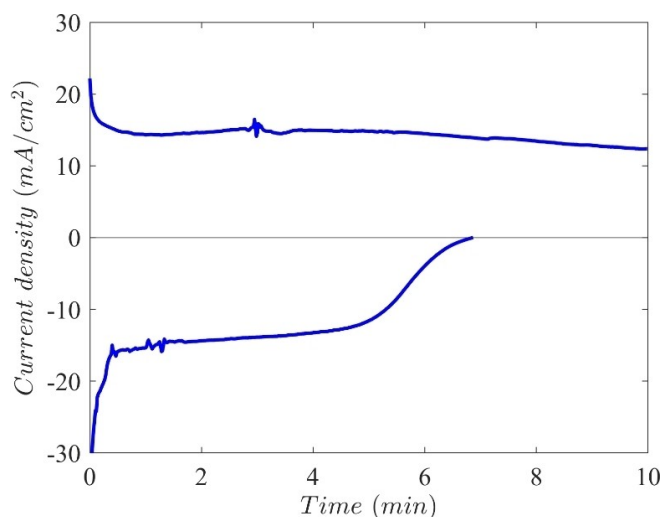
The slow kinetics of vanadium electrolyte, and the high self-discharge favored by high electrolyte concentration (1.8 M), leads to small values of coulombic efficiencies, resulting in a system that, as expected, is not efficient enough. As a result, the research efforts presented in this work shifted to the use of other electrolyte with faster kinetics, based on quinones.

Membraneless Alkaline Quinone Micro Redox Flow Battery (MAQMRFB): Anthraquinone and Ferrocyanide

Vanadium electrolyte is commonly employed in commercial redox flow batteries due to its unique properties. One advantage is the capability to mix the positive and negative electrolyte tanks, achieving a 50% V^{4+} and 50% V^{3+} mixture, allowing a simple rebalancing of the battery when capacity fades, a feature not shared by other electrolytes. Consequently, the use of alternative chemistries to Vanadium presents a great challenge in membraneless micro redox flow batteries, as the crossover of species is inevitable. When the membraneless micro redox flow battery starts its operation, it requires time for the fluid control system to actuate and stabilize the interface. This induces mixing of the positive and negative streams at least in the initial stages of operation. Hence, a decision was made to employ an identical electrolyte in both the positive and negative streams, composed of potassium ferrocyanide (Fe^{2+}, 0.2 M) and 2,6-dihydroxyanthraquinone (AQ, 0.06 M) in alkaline solution (KOH 1 M).[28,29] This approach entails the utilization of the previously described membraneless micro redox flow battery operating with this new electrolyte. For its operation 300 μL were fed into each stream (positive and negative).

Figure 6 illustrates the current obtained during the charging and discharging process of the new electrolyte in a membraneless micro redox flow battery, with constant flow rates of 250 μL/min and 200 μL/min for the positive and negative sides, respectively. Like MVMRFB, electrochemical experiments are conducted in potentiostatic mode (applying a constant voltage). In the charging case, a voltage of 1.4 V is applied for 10 minutes, and discharge occurs at 0.7 V, for the duration necessary to achieve an approximate zero current, indicating complete discharge. This approach allows to perform charge and discharge cycles of the membraneless micro redox flow battery previously presented, but now using a mixture of AQ and Fe^{2+} as an identical electrolyte, operating in closed fluidic loop, and with closed-control loop integrated. A coulombic efficiency of 57.85% with a capacity of 1.14 Ah/L (32.37% of theoretical electrolyte utilization) is obtained for the first cycle. It is worth noting that, even when electrolyte concentration is reduced by one order of magnitude, capacity is almost 30% from the one obtained with Vanadium and coulombic efficiency has significantly improved from values around 10–20% for the case of MVMRFB to values around 20–60% in MAQMRFB.

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Figure 6. Current profile obtained for the first cycle of the membraneless micro redox flow battery with Fe^{2+} and AQ as electrolyte in alkaline media. 1.4 V and 0.7 V were applied for charge and discharge processes, respectively.

Following the first cycle, due to implementation of the microfluidic control system, it has become possible, for the first time in membraneless micro redox flow batteries, to operate with organic molecules and study their cyclability. Figure 7 depicts the discharge capacity obtained for up to 60 consecutive cycles of charge-discharge for the MAQMRFB, along with the respective coulombic efficiency.

For the sixty cycles shown, a capacity loss of 1.18% per cycle is achieved based on the initial capacity value, being lower than for MVMRFB. In addition, for the coulombic efficiency, the loss is only 0.63% per cycle. It should be mentioned that the average current for the charging process is 3.3 mA (13.4 mA/cm²), and for the discharging process is 2.9 mA (11.7 mA/cm²), meaning that the power obtained in the discharge process is 9.15 mW/cm². The detailed information regarding current, power, energy, electrolyte utilization, and
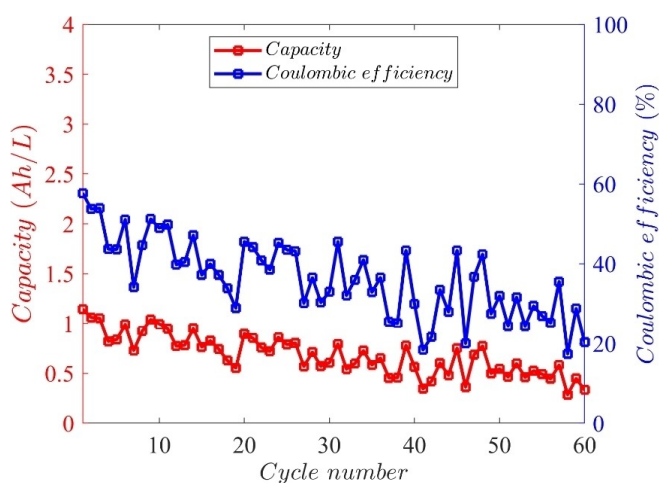
Figure 7. Cyclability results of the membraneless micro redox flow battery with Fe^{2+} and AQ as electrolyte.

efficiencies (energy and coulombic) of six relevant cycles (1st, 10th, 20th, 30th, 40th, 50th, 60th) from the experiment, is shown in Table S2 of Section S3 of Supporting Information.

Membraneless Micro Redox Flow Batteries Metrics

As discussed before, this is the first time that a charge-discharge process is achieved in a membraneless micro redox flow battery operating in a closed fluidic loop in the same way as conventional redox flow batteries. Table 1 compares membraneless micro redox flow batteries presented here, both for Vanadium and organic alkaline electrolytes, with previous literature. It is worth mentioning that, since there is no cycling data published before, only one charge-discharge cycle will be shown. For MVMRFB, the results of the first charge-discharge process tracked via spectroscopy are displayed, and for MAQMRFB first charge-discharge cycle is displayed. Normalization has been selected to compare the same geometric area in all the cases, the projected area of the electrode.

For the Vanadium electrolyte, some relevant results are the increase in discharge current density by an order of magnitude with respect to the one obtained by Lee et al.,[7] while volumetric energy density and volumetric capacity are multiplied by 6.4 and 4 respectively. Due to the operation in recirculation mode, around 50% electrolyte utilization is achieved via high flow rates in comparison to Lee et al.[7] However, multiple passes through the micro reactor increase self-discharge while decreasing coulombic efficiency, and that, together with a wider range of operating voltage, will impact energy efficiency of the MVMRFB presented here.

For the Alkaline Quinone electrolyte, the comparison is more complex since valid metrics are not available. In fact, to the knowledge of the authors, there are only three publications that operate membraneless micro devices with organic redox species.[8,18,19] Two of them are from the last two years, one of them operates only in fuel cell mode[19] and the other operates in recirculation mode, but this second one could not be considered a battery due to the same reaction taking place in the positive and negative half cells which yields a net zero voltage.[8] In any case, the performance obtained from the MAQMRFB described in this study outshined those of previous publications, especially in terms of energy efficiency. This is the first time that a membraneless micro redox flow battery with organic redox species achieves energy efficiency. Furthermore, it is the highest energy efficiency ever obtained until now, even when compared with Vanadium electrolyte.

Conclusions

The present study demonstrates, for the first time, a charge-discharge process with multiple-cycle operation of a membraneless micro redox flow battery, not only with commercial Vanadium electrolyte, but also with Alkaline Quinone electrolyte. The device is fully integrated by micro actuators, micro sensors, a closed-loop control system, and operation is carried

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doi.org/10.1002/batt.202400331

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Table 1. Comparing State-of-the-Art metrics for Membraneless Micro Redox Flow Batteries.

	Lee et al.[7][a]	B5TEC MVMRFB	Chaabene et al.[19][b]	Park et al.[8][c]	B5TEC MAQMRFB
Concentration (M)	1	1.8	0.1–0.35	0.01	0.06–0.2
Discharge Current (mAcm^{-2})	2.9	30	0.65	0.5	12
Discharge (mAh)	0.32	8.3	–	0.006	0.34
Discharge Power (mWcm^{-2})	2.9	24	0.04	0.071	8.43
Discharge Energy (WhL^{-1})	5.8	16.5	–	0.001	0.96
Discharge Capacity (AhL^{-1})	5.8	21	–	0.01	1.14
Electrolyte utilization (%)	20	43	–	33.8	32
Coulombic Efficiency (%)	34	21	–	66.4	58
Energy Efficiency (%)	20	10	–	–	29

[a] This experiment was carried out with non-continuous recirculation of electrolyte. Only one-pass of the electrolyte through the cell for the charge and discharge. [b] This experiment was carried out in fuel-cell mode, only a discharge of the electrolyte was made. [c] This experiment is in a symmetric cell, using a galvanostatic charge-discharge technique, since the reaction taking place in the negative and positive electrolyte is the same the net voltage is zero.

out in electrolyte recirculation mode, as conventional redox flow batteries, for the charge and the discharge.

A characterization of the micro reactor operating with Vanadium has been carried out through polarization curves at different flow rates and compared with previous results in literature. It is demonstrated that performance of the reactor involves the highest power and current densities achieved until now without involving modifications of electrolyte, only with an improvement in electrical external contacts which reduce contact resistance. With the improved micro reactor and developed microfluidic control system the first charge-discharge process is performed. A discharge capacity of 8.3 mAh, which corresponds to 21 Ah/L for a mean discharge current density of 30 mA/cm2, a coulombic efficiency of 21%, and an electrolyte utilization of 43% are observed. Through real time tracking of State of Charge by UV-visible spectroscopy using micro-optical cells, a maximum of 60% State of Charge is demonstrated after 4 hours of operation.

For the membraneless Vanadium micro redox flow battery eleven continuous cycles were performed, with an initial capacity of 4.2 Ah/L, and coulombic efficiency of 11.67% for the first cycle. Successive cycles present a gradual decrease in capacity, around 7.8%, while coulombic efficiency only decreases 0.5% per cycle. Several issues arised due to Vanadium's slow kinetics, its significant self-discharge, which reduces performance due to its high concentration, and the side reactions for high values of State of Charge. These challenges have led to the introduction of a new alkaline quinone electrolyte to operate the membraneless micro redox flow battery in the research presented here.

For the membraneless Alkaline Quinone micro redox flow battery sixty continuous cycles were performed, with an initial capacity of 1.14 Ah/L, and a coulombic efficiency of 57.85% for the first cycle. Successive cycles present a gradual decrease in capacity around 1.2% while coulombic efficiency only decreases 0.63% per cycle. The capacity of the battery decreases with

respect to its Vanadium analogue, but the rest of metrics demonstrate a significant improvement.

Finally, metrics of the MVMRFB and MAQMRFB presented in this study are compared with previous state-of-the-art. It is shown that the membraneless micro redox flow batteries presented here definitely surpass performance of previous publications. Furthermore, in contrast to those previous results, the ones presented here allow operation in recirculation mode like conventional redox flow batteries. The complete device is exclusively integrated by micro components, resulting in a membraneless micro redox flow battery that fits in the palm of a hand. The next step would be to generate a battery management system including, not only the classical power management unit, but also an interphase and fluidic management unit to automate and improve the performance. Nevertheless, here we present the first proof-of-concept of a membraneless micro redox flow battery.

Supporting Information

The authors have cited additional references within the Supporting Information.[30]

Acknowledgements

Conflict of Interests

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available in the supplementary material of this article.

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