

Redox flow batteries for energy storage: their promise, achievements and challenges

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Abstract

Redox flow batteries continue to be developed for utility-scale energy storage applications. Progress on standardisation, safety and recycling regulations as well as financing has helped to improve their commercialisation. The technical progress of redox flow batteries has not considered adequately the significance of electrolyte flow velocity, mass transfer and plug flow reactor modelling, despite steps in the right direction. 3D simulations of fluid flow, pressure drop, current distribution and mechanical resistance using commercial software are becoming more common, but satisfactory validation by experiments is still unusual. The majority of research tends to report short term studies on small electrodes, often in poorly defined flow channels; long term evaluation of electrode and membrane durability on a pilot scale is needed. Digital imaging of electrode structure using X-ray computed tomography is increasingly being used. Much activity is directed to organic and non-aqueous systems. However, scale-up and high, sustained charge capacity using electrolytes of moderate cost which are environmentally acceptable to source, store, transport and handle require considerable attention. Recommendations for future work are discussed.

Keywords: electrochemical engineering, energy storage, mathematical modelling, redox flow batteries, state of charge, vanadium

(2,400 words; 4 figures; 97 references, 14 of them commented on)

Introduction

The deployment of redox flow batteries (RFBs) has grown steadily due to their versatility, increasing standardisation and recent grid-level energy storage installations [1]. In contrast to conventional batteries, RFBs can provide multiple service functions, such as peak shaving and sub-second response for frequency and voltage regulation, for either wind or solar power generation and the power grid. Relevantly, they are now considered a safer alternative to Li-ion stationary systems for indoor use in buildings [2]. Since devices complying with local legislations on fire safety and electrolyte management will have competitive advantages, standardisation of RFBs has been progressed via the International Electrotechnical Commission [3], as an addition to updated grid storage standards [4].

The large-scale deployment of RFBs in a multi-device energy market with many service providers has been hindered by the perception that the technology is still in an early stage of development and by the relatively high capital costs due to electrolytes (e.g. vanadium) and ion exchange membranes. However, market conditions have become more favourable for RFB manufacturers. The rising cost of wind power curtailment has incentivised heavy investment in China [5], while electrolyte leasing models have relieved the impact of vanadium costs [6]. Moreover, the price of cobalt in Li-ion cell cathodes has more than doubled since 2016 [7], due to the ‘high-risk’ status of its supply chain [8]. Work continues on the commercialisation of other RFBs systems, mainly zinc-bromine [9] and the search for feasible organic chemistries continues [10]. The prospective success of such systems could overcome present limitations. While the principles of this technology are well-known, the path to scale-up and development has often been unclear to researchers [11]. The full potential of RFBs has yet to be realised. Progress is currently

hindered by the scarcity of electrochemical engineering studies on reaction environment and cell design and their relation to performance and efficiency.

Mass transfer in redox flow batteries

RFBs consist on arrays of electrochemical flow reactors [11]. Reactant conversion, cycling performance and energy efficiency are related to the global mass transfer coefficient, k_m , due to the mixed reaction control at each electrode, which depends on the supply of reactant. The electrochemical performance is always tied to the total active electrode surface area, A , which can be difficult to determine and can even be time-dependent (mainly during the initial commissioning). A simple method to characterise electrochemical cells under full mass transfer control is to obtain the performance factor k_mA , typically for a diluted model redox couple. In porous electrodes, the surface area per unit volume, A_e , can be considered, the factor thus becoming k_mA_e . The fundamental relationship to the limiting current, I_L , is:

$$k_mA_e = \frac{I_L}{zFcV_e} \quad (1)$$

where z is the electron stoichiometry, F is the Faraday constant, c is concentration and V_e is the volume of the electrode. The performance factor can predict the rate and fractional conversion (maximum rate of the state of charge, SOC) during scale-up using either analytical or numerical modelling approaches. Empirical power laws are also instructive, e.g., k_mA_e as a function of mean electrolyte flow rate, v , as well as dimensionless correlations involving Sherwood, Péclet, and Schmidt numbers, which facilitate scale-up and comparison among different cells. Further work can extend the implementation of the fundamental expression when considering significant single-pass conversion, concentrated electrolytes or parasitic reactions.

A classical route to determine $k_m A_e$ is the measurement of limiting currents assuming a steady state, as applied recently to electrode materials within rectangular channel flow-through cells for cerium-based RFBs [12], and borohydride fuel cells [13]. This factor can be related to the pressure drop, ΔP , over a range of v values, as shown in Fig. 1, as an indication of scale-up feasibility [14]. Each correlation in the plot corresponds to the behaviour of a different electrode material in a particular cell compared to the FM01-LC electrolyser. A modified expression relating $k_m A_e$ to a limiting current has been experimentally validated for the case of carbon felt electrodes using the $\text{Fe}^{2+}/\text{Fe}^{3+}$ redox couple in a diluted electrolyte [15]. Meanwhile, values for k_m at the pore level have been obtained in a similar method using vanadium electrolyte in a laboratory RFB [16].

A way to increase mass transfer is the use of a zero-gap electrode architecture with flow field designs [17,18,19], which have been widely used in gaseous fuel cells. This strategy has already demonstrated significant improvements to the power density of vanadium cells and stacks [20], reaching values up to 2588 mW cm^{-2} [19]. (Challenges to their implementation are discussed in a following section.) The mass transfer properties of cells having flow fields, however, have received insufficient attention. One of the first works towards establishing k_m for these features was carried out with vanadium electrolyte and carbon felt via the limiting current method [16]. As shown in Fig. 2, $k_m A_e$ was later determined for different flow field designs in a single-electrolyte flow cell using carbon paper as a function of v at the flow channels using a one-dimensional model [21]; a step towards reporting normalised flow in interdigitated cells. Dimensionless performance parameters showed that interdigitated flow fields outperformed other designs. In a

following work [22], the same group evaluated the effect of flow rate and electrolyte viscosity on $k_m A_e$, limiting current and pressure drop. An expression for k_m considering the viscosity of the electrolyte was proposed:

$$Sh \equiv \frac{k_m d_f}{D} = \frac{Da \cdot \kappa_{eff} R T d_f}{\theta F^2 c_{A_e} S^2 D} \quad (2)$$

Where Sh is the Sherwood number, d_f is the fibre diameter, D is the diffusion coefficient, Da is the so-called Damköhler number, κ_{eff} is the effective solution conductivity, R is the universal gas constant, T is temperature and S is the electrode thickness. Notably, the exchange current density to limiting current density ratio, θ , an often-unappreciated ratio for electrode processes, is contained in the equation and was plotted as a function of v .

The pressure drop over flow fields in cells [23,24,25], has yet to be directly correlated to mass transfer. In some cases (e.g. certain laboratory-scale cell suppliers) flow field designs for proton-exchange fuel cells (PEMFCs) have simply been moved across to RFBs with insufficient thought. PEMFCs employ gases, while most RFBs involve liquids with much higher viscosity. Furthermore, the transport properties of the flow fields depend on their size and shape and do not scale-up linearly [24]. Thus, efforts in this promising area should be directed towards studies which can be readily scaled-up or represent full-size cells, e.g. [26].

Both the local mass transfer at fibre level and the global mass transfer coefficient in utility-scale electrodes experiencing non-homogeneous porous fluid flow can be simulated. For instance, by digitising their geometry from X-ray CT scans, mass transfer coefficients and dimensionless correlations have been calculated for carbon felts using

the Lattice Boltzmann method (LBM) [27,28]. This allowed correlations to be established at low Reynolds numbers [27], along global dispersion and reaction rate coefficients [28]. The local mass transfer at felts within interdigitated flow fields has also been determined by numerical simulations [29,30], although those for flow-through rectangular channel electrodes have not received adequate attention. Contrary to the previous examples, the models discussed in the following section emphasise other parameters and incorporate mass transfer rates from reported correlations. It should be noted that any employed correlations must be strictly valid for the model assumptions.

Mathematical modelling

In general, the modelling of RFBs is accompanied by insufficient experimental validation and the introduction of arbitrary fitting factors. Numerous examples exist of practical zero-, one- and two- dimensional models of cell potential over charge-discharge cycles. However, given the widespread availability of computational power, opportunities for improvement can be found in the replacement of simplified assumptions for representations of the physical geometry of, and reaction environment within, the device, e.g., dispersed plug flow reactor (PFR) models applied to three-dimensional electrodes. Models considering PFRs are scarce [31,32], despite flow reactors generally following this behaviour. There is also a tendency to omit reference electrodes from each half-cell, ignore the value of a redox probe in the reservoirs and neglect the classical model of a PFR with batch recirculation to a fixed volume electrolyte reservoir (for mass transfer-controlled cases, which are useful for reactor design and characterisation).

Zero-dimensional models for flow rate optimisation have been recently considered [33]. For instance, an iterative model has been used to determine a variable flow factor as a function of SOC and coupled to pressure drop [34]. The model follows a quasi-plug flow reactor as a compromise with computational efficiency [32]. The concentration overpotential in a commercial vanadium RFB was later modelled using a zero-dimensional approach [35]. Since this type of model cannot consider the porous area of the electrode, a scaling factor was applied to the geometrical electrodes in order to produce practical results. Other RFBs models develop control algorithms, e.g., extended Kalman filter (EKF) for capacity monitoring [36], SOC estimation considering the capacity fading [37], and time flow control for charging using solar cells [38] or wind power [39].

Techno-economic feasibility models have been recently reviewed [40]; most of them centre on capital cost, stressing the significant role of vanadium reactants, and revealing wide variability in the assumed prices of membranes and ancillary equipment. Recently, two models for grid-level energy storage, one in Germany [41], and the other in the USA [42], concluded that RFB investment costs should be reduced by 20-30%, in order to remain competitive. Notably, a model for the implementation of large-area electrodes has estimated potential cost reductions of about 25% [43], while new models could establish up-to-date cost and financing targets by taking into account new electrolyte leasing schemes [6]. Regarding lobbying opportunities, a case study has pointed out the need for flexible tariffs in order to maximise storage value [44]. The environmental life cycle assessment of vanadium systems has been carried out [45], highlighting their improved recyclability compared to Li-ion batteries.

Numerical, three-dimensional ‘multiphysics’ simulations can evaluate the fluid flow distribution, mass transfer-driven current density, pressure drop and shunt currents over the whole electrode and cell geometries while describing charge-discharge performance. Such models have permitted to analyse the efficiency of different stack configurations [46] and cell designs [47]. Other models have focused on specific parameters, such as flow rate and pressure trade-offs [48,49,50], or electrode polarisation and contact resistance due to felt compression [51]. Recent mechanical models of cell stacks [52] should aid reliable manufacture and durability.

A variety of cell designs has been studied in terms on CFD fluid flow and pressure drop calculations, including flow-through cells [53], as well as serpentine and interdigitated flow fields [24,49,54,55]. Interdigitated flow fields can be more effective in ensuring high mass transfer. Necessary optimisation studies of the dimensions of flow field channel depth and width, in relation to the bipolar plate ‘rib’ area, are available for bromine [29], and vanadium cells [30]. Normalised, validated scale-up for flow field designs provides an important research opportunity. A marked limitation of these studies is that they consider cells under 10 cm² electrode area, whereas utility-scale cells might involve 2,000 cm² electrodes. As mentioned before, current correlations applied to flow fields scale-up in a non-linear manner.

Current and potential distribution are the result of the combined effect of electrical resistances, reaction kinetics, mass transfer and electrode geometry. A validated model for such phenomena in interdigitated flow fields has been developed [56], considering non-uniform compression of the felt [57]. Experimental current distribution studies have

shown non-homogeneous patterns in some interdigitated flow fields [58], a concern that must be solved for successful scale-up. Primary, secondary, tertiary or *actual* current distributions, as well as the method for coupling mass transfer to the tertiary distribution are missing from these studies. On the other hand, validation of potential distribution during mass transfer limiting conditions has been achieved using platinum probes [59].

Numerical simulations have addressed shunt currents in cell stacks. For instance, in conjunction to electrode overpotentials and manifold ionic current [60], and in terms of electrochemical reaction rate distribution [61]. As shown in Fig. 3, the latter study highlights that localised corrosion due to opposite, parasitic electrode reactions can take place near electrolyte manifolds if the stack does not incorporate anti-shunt currents features. The trade-off of shunt current distribution has also been estimated for different stack configurations [46]. In previous studies, scenarios that allow shunt currents have been deliberately analysed. Effective preventing measures must also be modelled. Equivalent circuits of shunt currents in utility-scale devices continue to be an effective tool to predict losses and optimise the number of cells [62].

Manufacturing, materials and utility-scale research needs

The manufacture of RFBs aims to achieve reliability over extended life-time, dimensional tolerances for scale-up and cost-effective mass production. Few works have focused on the longevity of materials. For instance, degradation of proposed polypropylene [63] and sulfonated membranes [64], or aging of carbon felt [65,66,67]. Regarding manufacturing tolerances, stacks under compression are now being evaluated using X-ray computed tomography (CT) [68], revealing possible deformation after assembly or over time. This

approach could be used to validate mechanical stress distribution models [52]. CT is also being employed to study the effect of compression on carbon felt properties [27], and to determine the surface area of porous electrodes (but only after calibration and validation of the methodology) [69]. In relation to materials research, limited attention has been given to the advantages and limitations of composite membranes [70,71] and to the homogeneity of carbon felt, which is important in large cells.

Considerations such as electrolyte fluid flow, mass transfer, pressure drop, active electrode surface area and current distribution at relatively large electrodes become important at a realistic scale [11]. Hence the need to obtain data from larger cells which experience edge and manifold effects, flow maldistribution, and shunt currents, all under power conditioning regimes [35,72,73,74,75]. If needed, practical characterisations of such systems can be carried out in a cost-effective manner. This has been demonstrated for a utility-scale vanadium RFB, shown in Fig. 4, which was studied in energy efficiency terms (accounting for convertor efficiency, parasitic reactions and pumping demands) by using accurate transducers and instrumentation.

Meanwhile, the development of extended-area, flexible 2.5 m² electrodes based on welded carbon felt-sheets has been announced [76]. Mathematical models have shown that such an intensive scale-up could bring down RFBs capital costs significantly [43]. The design of large cells could benefit from experience in chlor-alkali stacks, although it will be essential to study and report the effects of flow dispersion, pump energy demand and concentration gradients on their efficiency. Regarding the development of bipolar plates, the requirements for the manufacture of flow fields should be discussed, together

with their impact on the cost and complexity of stacks. These features are typically made by Computer Numerical Control (CNC) manufacturing, but it is not clear if they could be produced by alternative methods, e.g., injection moulding. The possibility of blockage by fibre sludge in large cells over extended cycling should be considered and prevented. Additive manufacturing to build 3D objects has also been considered to design electrochemical cells with characteristics that would not be possible to realise by CNC and to create rapid prototypes [11].

Research should also pay more attention to the experimental characterisation of pressure drop [77,14,78], in special due to the urgent need for validation of CFD models. As mentioned previously, simulations have frequently neglected comparisons against experimental data. Still, the optimisation of pumping power consumption is increasingly analysed [79,77], especially for interdigitated flow fields [49,22], although intrinsically more efficient pumps have yet to be considered [80]. Pulsating flow changing its direction has been visualised in laboratory cells relying on peristaltic pumps [49]. This is the reason why pulse dampeners should be implemented to achieve continuous flow in such cells [12]. Flow visualisation can incorporate thermal imaging [49,81].

Regarding other practical issues, water transfer through the membrane, the effect of migration and the necessary system rebalancing to recover capacity fading in vanadium RFBs are increasingly acknowledged [82,37,36,83]. However, detailed balancing procedures for large cells are practically missing from the literature. Despite the availability of many laboratory methods, reliable plant-level SOC monitoring is still needed, as commercial systems use charge integration due to the inconsistencies in

monitoring cells after cross-mixing over many cycles. To address this, extended Kalman filters have been adopted [36,37]. Investigations continue regarding *in situ* current distribution and single pass SOC changes [84]. On a different matter, problems with the reproducibility of the composition and purity of vanadium electrolytes have been identified [85]. Indeed, best practice and standard procedures for the chemical analysis of vanadium electrolytes have not been agreed.

Organic and other alternative chemistries

Organic electroactive species dissolved in aqueous or non-aqueous media represent an attractive alternative to inorganic redox species. Organic molecules can be manipulated by introducing functional groups that can alter their solubility, redox potential [86], energy density [87], or choice of supporting electrolyte [88,89]. Recent review papers [10,90], have highlighted the challenges for developing organic redox flow batteries which include: appropriate materials, safety, low cost, high energy density, larger electrolyte volumes, symmetrical and unsymmetrical cyclability, development of mathematical models and the design of stacks with the appropriate electrochemical engineering. Typical electrolytes for organic RFB are non-aqueous and, more recently, eutectic electrolytes [91]. Most organic chemistries are still challenged by poor stability, hygroscopicity and difficult or expensive synthesis. Work has indeed focused on the stability of redox organics [92,93], including disproportionation systems [94,95], which are interesting for having the same initial composition in both half-cells. Ongoing research should be directed towards demonstrating the reliability of organic RFBs beyond the glovebox (low oxygen, dry atmosphere), at the laboratory pilot-scale. This will require the suitability of the components and materials employed in the flow system to be scrutinised, together with the need for ancillary dry or inert gas supplies, all of which

must be cost-effective. Of course, these complications could be avoided by finding inexpensive, stable and easy-to-handle organic redox couples and solvents.

Conclusion

Several general points may be made:

1. The majority of academic studies focus on the development of catalysts tested in small electrochemical cells. However, they are not realistic for advancing the technology of RFB; they are limited to short term laboratory experiments and, in general, the figures of merit such as mass transport, space time velocity or pressure drop suitable for scale-up, are not reported. The durability of the stack, catalyst and electrode materials still receive insufficient attention.
2. The reaction environment in many cells remains unclear, due to inadequate attention to experimental measurements on the distribution of current, potential, concentration and flow velocity; flow visualisation studies are rarely reported; on-line determination of accurate SOC is still under development.
3. Simplified computational models tend to be used, being validated in fewer cases than expected. 3D models extended the scope beyond flow dynamics into the assessment of current distribution, mechanical considerations and shunt currents.
4. The use of small electrodes and cells in poorly defined flow channels does not provide current-electrode area data which will scale-up in a linear fashion, due to maldistribution of current and flow. Examples of studies employing representative electrode sizes are found in references [96] and [26].

5. Many studies do not take the opportunity to incorporate reference electrodes in each half-cell, e.g. see reference 97, enabling potential distribution and cell potential components to be expressed as a function of current density for each electrode reaction.
6. Few studies of vanadium RFBs appreciate that OCP probes in the half-cell electrolyte reservoirs or flow loops can be used to monitor SOC via the solution redox potential.
7. It is poorly understood that electrolyte flow regime, channel equivalent (hydraulic) diameter, mean linear flow velocity of electrolyte and pressure drop are critical parameters characterising any RFB flow cell.
8. Regardless of the type of study, the mass transport properties of a RFB cell should be assessed as a function of mean electrolyte velocity past the electrode surface, v , e.g., against a diluted model redox couple. This would provide a basic quantitative indicator of their reaction environment and permit more rigorous comparisons against other experimental conditions and cell or stack designs/sizes.
9. There are very few case studies of scale-up, full size cell characterisation or plant performance during extended operation. While proprietary considerations are appreciated, this situation limits confidence in future roll out and wider deployment of the technology.

Conflict of interest statement

The authors declare that there is no conflict of interests.

Acknowledgements

LFA and CPL acknowledge the financial support of Newton Fund and Innovate UK. LFA acknowledges additional support from the RIFI of the University of Southampton and fruitful discussions with colleagues at conferences.

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Figure Captions

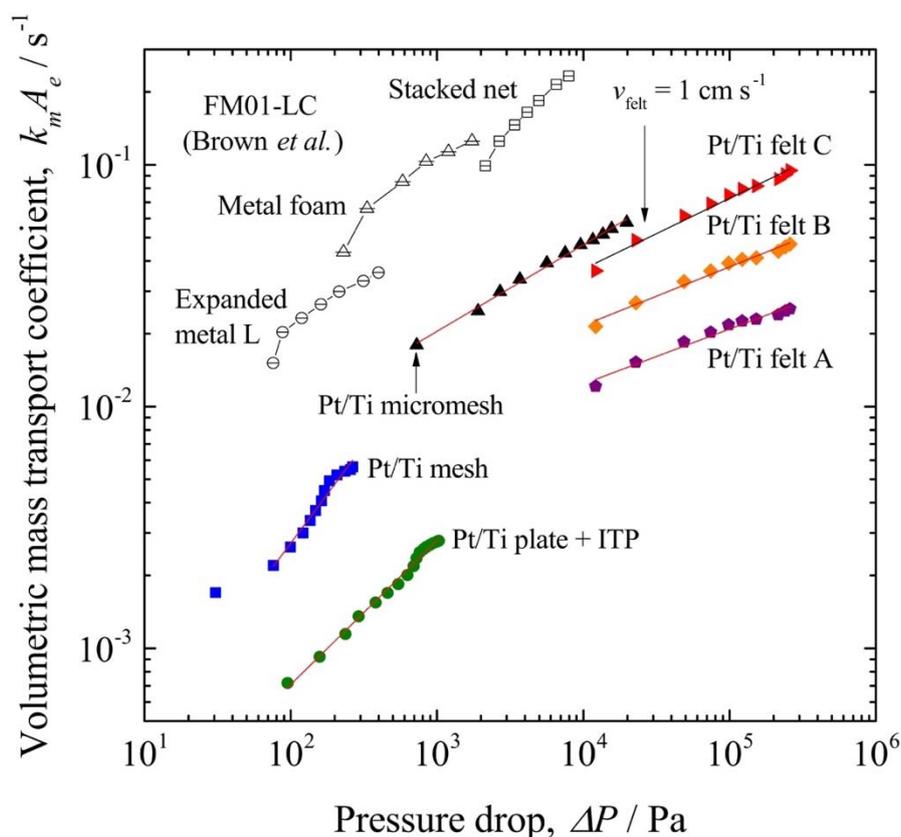


Figure 1. The normalised performance of two cell designs under full mass transfer reaction control having different porous electrodes and electrolytes. The product $k_m A_e$ is plotted as a function of pressure drop due to electrolyte flow rate. Pt/Ti electrodes correspond to a laboratory cerium-based RFB and others to a FM01-LC electrolyser [12]. Reprinted with permission from Arenas *et al.* [14], Pressure drop through platinised titanium porous electrodes for cerium-based redox flow batteries, *AIChE Journal* 64(3):1135–46. Copyright (2017) John Wiley and Sons.

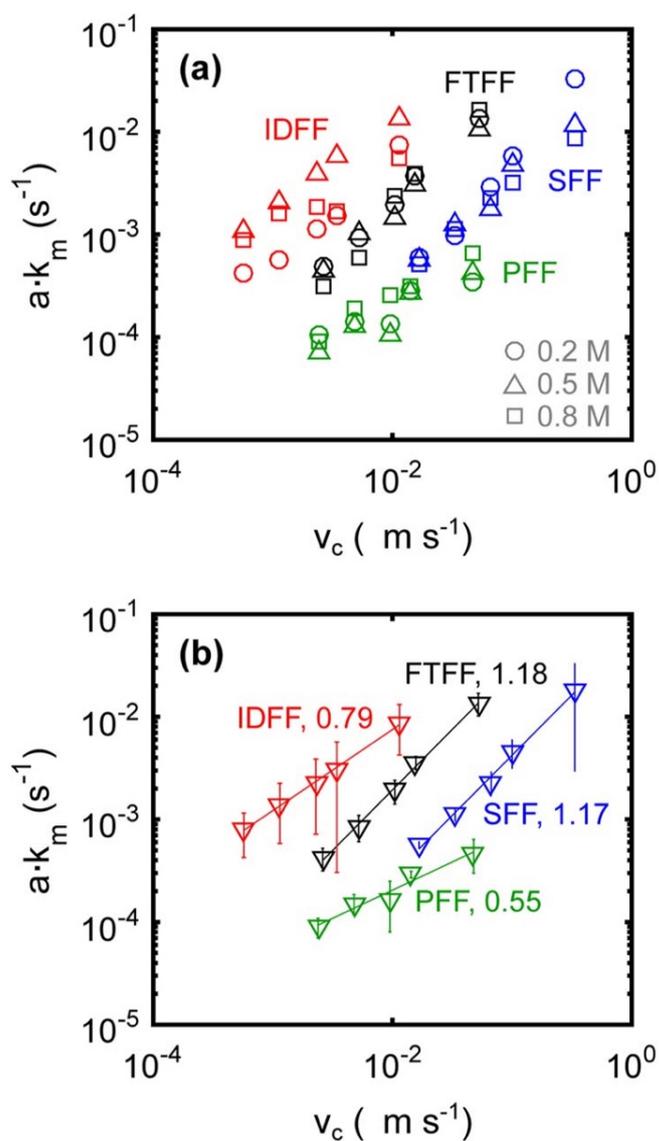


Figure 2. The performance of different flow field designs in a 2.55 cm² symmetrical cell for the Fe²⁺/Fe³⁺ redox couple. a) $k_m A_e$ is plotted as a function of characteristic flow velocity (related to flow field dimensions) for different concentrations: 0.2 mol dm⁻³ (○), 0.5 mol dm⁻³ (△), and 0.8 mol dm⁻³ (□). b) Mean $k_m A_e$ values fitted to linear relationships along their slope values. Flow field types: parallel (PFF), serpentine (SFF), interdigitated (IDFF), and flow through (FTFF). Adapted with permission from Milshtein *et al.* [21], Quantifying mass transfer rates in redox flow batteries, Journal of the Electrochemical Society 164(11):E3265–75. Open access licence (2017) CC BY 4.0.

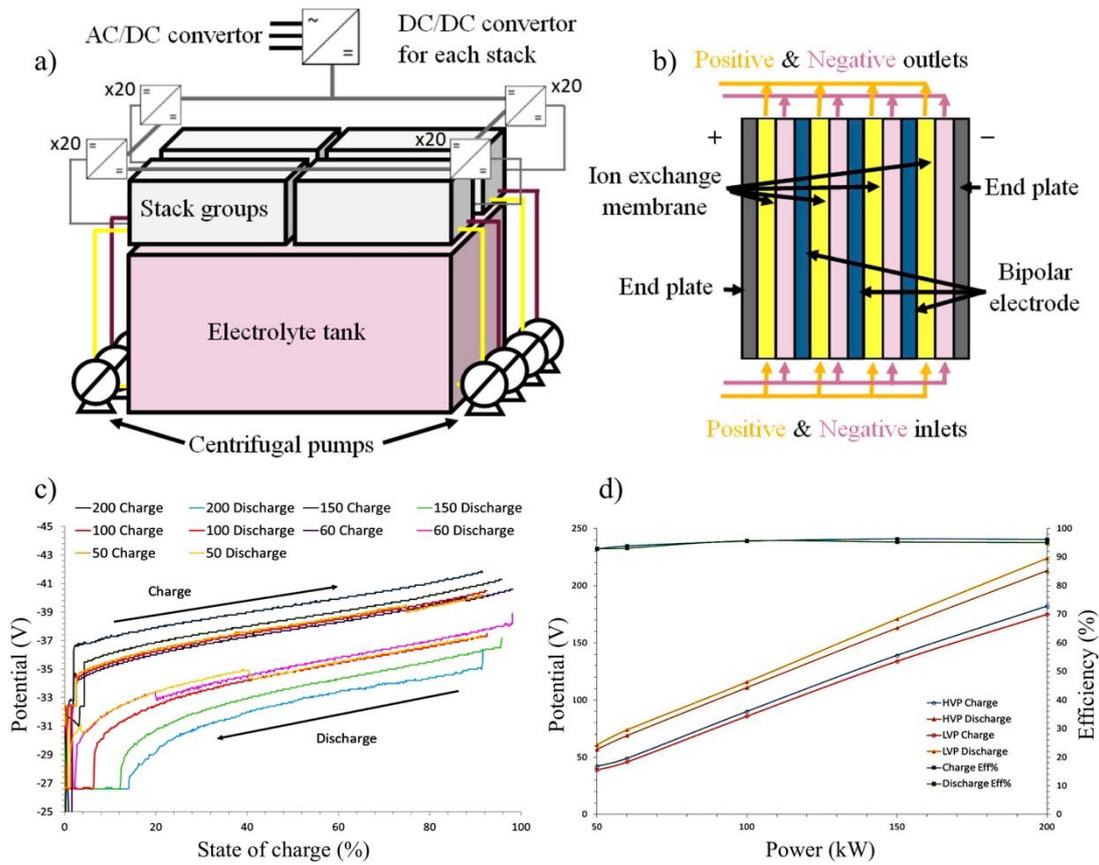


Figure 4. a) A four-stack 200 kW/400 kWh vanadium RFB along pump and power conditioner. b) Arrangement of bipolar electrodes and electrolyte flow. c) Total stack potential over a range of constant power charge-discharge values. d) DC/DC convertor potential losses and efficiency at ‘high voltage’ (HVP) and ‘low voltage’ (LVP) modes. Adapted with permission from Bryans *et al.* [75], Characterisation of a 200 kW/400 kWh vanadium redox flow battery, Batteries 4:54. Open access (2018) CC BY 4.0 licence.