Comparative Analysis for Various Redox Flow Batteries Chemistries using a Cost Performance Model

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Abstract:

The total energy storage system cost is determined by means of a robust performance-based cost model for multiple flow battery chemistries. Systems aspects such as shunt current losses, pumping losses and various flow patterns through electrodes are accounted for. The system cost minimizing objective function determines stack design by optimizing the state of charge operating range, along with current density and current-normalized flow. The model cost estimates are validated using 2-kW stack performance data for the same size electrodes and operating conditions. Using our validated tool, it has been demonstrated that an optimized all-vanadium system has an estimated system cost of $< \$350\text{kWh}^{-1}$ for 4-h application. With an anticipated decrease in component costs facilitated by economies of scale from larger production volumes, coupled with performance improvements enabled by technology development, the system cost is expected to decrease to 160 kWh^{-1} for a 4-h application, and to $\$100\text{ kWh}^{-1}$ for a 10-h application. This tool has been shared with the redox flow battery community to enable cost estimation using their stack data and guide future direction.

Key words: cost, performance, flow, interdigitated, energy, power

Introduction

Renewable generation worldwide has grown from 220 TWh of generation in 2009 to 446 TWh in 2011, at a compounded annual growth rate (CAGR) of 29.6% [1, 2]. The global wind installed capacity increased from 55 GW to 275 GW in the 2005 to 2012 timeframe, corresponding to a CAGR of 26% [3]. From May 2011 to December 2013, the amount of energy storage excluding pumped hydro and compressed air nearly doubled [4].

The catalysts for this growth are mandates for a minimum amount of electricity production from renewables in Europe, US and elsewhere. In Europe, the mandated energy production from renewables is 20% by the year 2020, which includes electricity, transportation, heating and cooling [5, 6]. In the US, mandates in several states have been in place since January 2012 [7], California leading the way with a requirement of 33% renewables generation by 2020. The wind installed capacity in China increased at a CAGR of 44% to 90 GW in 2013 from 10 GW in 2007. The US wind installation has grown at a 15% CAGR in the same period to 61 GW, double the growth rate of Germany in the same period [3]. Energy storage required for dispatchable power was estimated to be 20% of the nameplate capacity for wind and solar generation [8].

A recent study showed 18.6 GW/8.6 GWh of energy storage was required to provide balancing services to accommodate additional wind capacity and load growth [9], while arbitrage as a stand-alone application was not found to be cost effective. A demonstration project was completed by PNNL and its project partners to explore use of storage for multiple applications by prioritizing its use based on revenue potential, state of charge and grid-based criticality for a 120 kW/500 kWh Li-Ion battery system [10], while a similar effort is underway for a zinc bromine flow battery system [11].

There have been several demonstration projects for storage, with Li-ion battery systems used for power intensive application such as frequency regulation, and flow batteries and sodium sulfur batteries used for energy intensive applications. The largest Li-ion battery tested so far is an AES 32-MW, 8 MWh system using A123 batteries at Laurel Mountain in West Virginia for power regulation [12]. Plans are afoot for deployment of 2-4 hour Li-ion battery systems by AES [13]. The largest sodium sulfur battery is the 34 MW, 238 MWh storage system deployed at the Rokkasho-Futamata Wind Farm [13], while one of the largest planned redox flow battery systems is a 25 MW/75 MWh zinc bromine Primus Power system at Modesto, California irrigation district [4].

The California energy storage procurement requires utilities to procure a total of 1.325 GW of storage by 2022 [14], with additional procurements announced by New York, Hawaii, Washington, and Ontario, Canada [15]. To allow standardized testing of energy storage performance, the US DOE-OE sponsored an effort led by PNNL and Sandia National Laboratories to develop performance test protocols for various applications [16, 17]. This protocol effort has been adapted in its entirety by the performance working group (WG2) of IEC TC120 to develop international standards efforts for grid integrated electrical energy storage (IEC TC120), while a similar effort from the Electric Power Research Institute (EPRI) sponsored Energy Storage Integration Council (ESIC) is also relying on this document for performance related procedures. The DOE-OE test protocol has been test driven by multiple users and their feedback has been collected to identify gaps in the protocol. The EPRI-ESIC effort is further tailored towards identifying various use cases appropriate for grid-connected storage.

While conventional batteries have a fixed energy to power (E/P) ratio, flow batteries can vary their energy and power independently by varying the tank and stack size respectively. The wide

range of storage applications coupled with the impending growth in storage deployment leads to a critical need to reliably estimate the cost for the storage system for each application. High level studies have provided such estimates in the past [18, 19]. However, they do not take into account the actual performance of the system, the optimization of the operating parameters for each application, E/P ratio, stack design and balance of plant sizing (pump, heat exchangers). A detailed literature review of existing cost models and the gaps has been covered in our previous work [20] and reviews of redox flow battery systems can be found in numerous publications [21-24].

The earlier cost performance model developed by our team assumed the minimum system cost occurred at the maximum system operating efficiency [20]. However, this is not always the case. In this paper, the system cost objective function is minimized for various applications, giving rise to optimized stack design, balance of plant sizing and operating conditions such as current density, flow rate and SOC range corresponding to the minimum cost. In addition to conventional flow through the felt considered in our previous work for a 1-kW stack, this paper shows stack data for both conventional and interdigitated flow for a 2-kW stack, and explores the E/P ratios for which this flow pattern offers cost savings. The model was validated by comparing cost projection using performance data for the 2-kW 15-cell stack with the model cost projection for the same stack area and operating conditions – current density, flow rate and state of charge (SOC) range. Model validation allows use of this cost performance model to design and size the stack, size the pumps and determine the optimum operating conditions for various E/P ratios.

Model

The methodology has been described in detail in our previous work [20]. The stack or power module voltage curves was modeled by accounting for various losses such as ohmic,

charge transfer, electrolyte flow and shunt current induced losses. The optimum state of charge (SOC) operation range or depth of discharge (DOD) was determined for grid services with a wide range of energy requirements and a rated power of 1000 kW. 60-cell power modules were used for this analysis based on preliminary results that this provided a good tradeoff between shunting losses and power module costs. The cell components were sized based on the discharge performance at the low SOC end point for a given current density and flow rate per current. The flow rate at low SOC, being the highest, set the pump size. The current density and flow rate were adjusted to provide the required power across the SOC operating range.

The average discharge voltage across the SOC range was used to calculate the required electrolyte volume. Components costs are provided in [20], with N212 Nafion® membrane costs used in this work were estimated to be 45% of N117 Nafion® costs. The total system cost was calculated from the power and energy costs based on the Energy/Power (E/P) value. Fig. 1 shows the overall schematic for our approach.

The model was set up to analyze 3 chemistries; vanadium (V-V), iron vanadium (Fe-V), and iron chromium (Fe-Cr). The V-V system uses a mixed acid two molar electrolyte system developed by PNNL with high energy density and temperature stability [25]. The higher concentration requires lower flow rate, which reduces pumping losses and pump capital costs. The wider operating temperature range allows use of passive heat exchange and removes the need for a dedicated heat exchanger, while the smaller footprint is expected to decrease installation costs. In addition, in-house data for 0.4 kW and 2 kW scale stacks determined the pressure drop for the mixed acid electrolyte V-V system to be lower than the conventional sulfuric acid electrolyte. The lower viscosity for the mixed acid system allows use of higher flow rates without additional penalties associated with pressure drop losses. For the Fe-V system,

which has a lower operating voltage, the chloride supporting electrolyte enables higher active species concentration and wider temperature operating window, along with use of low cost microporous separator [26]. For the Fe-Cr system, the electrolyte viscosity was obtained from work done by United Technologies Corporation (UTC) [27] while performance curves were obtained at 54 mA cm^{-2} for a stoichiometric flow rate of 2 from Fig. 6 of [28]. These performance curves were similar to Fig. A-1 in the UTC work. While an ion exchange membrane was used by the above groups, the calculations assume a low cost microporous separator similar to the approach used for the Fe-V chemistry [20].

The model simulated the battery performance for various values of the following operating conditions: current density, temperature, current normalized flow rate, and SOC range. Available parameters such as electrode specific area, charge transfer kinetic rate constants, membrane conductivity, and porous separator tortuosity were incorporated. Fitting the model to the measured voltage vs. SOC curves at various current densities and flow rates allowed correction for the electrode thickness that actively participates in the charge transfer. Further details are available in our previous work [20] and also available in the GitHub, a web-based code repository [29]. For the all vanadium system, the kinetic rate constants from our previous work were updated with values provided by Yamamura et al [30]

A differential analysis was used to calculate the parasitic losses due to current shunting through the inter-cell electrolyte channels [20]. This method computes the current flowing through each manifold and flow channel, with subsequent calculation of the ohmic power loss associated with each flow path.

The electrolyte resistance in each flow channel can be adjusted by controlling the channel cross-sectional area. Higher area lends itself to lower electrolyte resistance, lowering flow related

inefficiencies while diverting more current flow through the inter-cell electrolyte path. Hence, an optimum design of flow channels is essential.

The shunting of current through the inter-cell electrolyte flow path increases with more series-connected cells [31]. However, reducing the magnitude of series-connected cells in a power module increases the cost related to end plates and inter-power module connections.

Darcy's law was used to calculate the pressure drop ΔP through each porous electrode [32] as described in our previous work [20]. The ratio of viscosity to permeability, μ/K , was found from stack data for V-V. The electrolyte viscosity for the V-V system was measured at PNNL across the SOC range for both the catholyte and anolyte. There was a slight variation (less than 4%) in the measured viscosity over the 10% to 90% SOC range for both electrolytes and between the two electrolytes at a fixed SOC. Using an average viscosity of 6.6 cP results in a corresponding permeability of $4 \times 10^{-10} \text{ m}^2$. The electrolyte viscosity was determined from stack ΔP measurements for the iron vanadium chemistry to be 9.5 cP, varying about 10% across the SOC range. For the Fe-Cr chemistry, a viscosity value of 2.2 cP was used from literature [27].

The pressure drop along the plastic frame flow channels were computed and added to the pressure drop across the electrodes, and the pumping work calculated as described in [20]. Data received from vendors was used for pump efficiency η . Note that pump efficiency is generally a function of flow rate ΔP . For the flow rates associated with the 1000 kW system, the pump performance curves provided by a vendor indicated a pump efficiency range of 65 to 86%, while a range of 65 to 74% was indicated in the literature [33]. The motor efficiency was 0.9 for power needs greater than 15 kW [34]. For this work, the product of pump and motor efficiency was assumed to be 60%. A list of symbols is provided in Table 1.

Power module component costs were shown in Table 1 of our previous work for three scenarios: present, near term and optimistic [20]. In our earlier work, N117 Nafion® membrane was used for the all vanadium system, while in this study, N212 Nafion® membrane was used. N212 costs were assumed to be 50% of N115 unit area costs, while N115 costs were assumed to be 90% of N117 unit area costs. A special case showing an order of magnitude cost reduction of the Nafion membrane with respect to present costs was also conducted based on Fig. 3-13 of Kamath *et al* [36].

Model deployment

The calculations were done for the discharge mode for a desired power and energy output, with the system consisting of thirty six 60-cell stacks [20]. A cost objective function was determined for a given power and energy requirement, as a function of the following operating parameters:

- current density at low SOC
- flow rate per current at low SOC
- SOC range.

Optimization was done by minimizing this cost objective function.

The three values that need to be determined in order to return a cost are:

1) the cell area to determine stack costs, 2) flow rate at low SOC to determine pump costs, and 3) effective voltage V_{eff} to determine electrolyte and tank costs. The cell area was calculated to provide the required power at low SOC, thus ensuring the ability to provide power at higher SOC's. The current density, flow rate per current, and low SOC are inputs to the cost function, which are optimized. Note that the low SOC fixes the DOD, as the SOC range of operation is assumed to be symmetrical. For applications that use longer charge duration, the

system can be charged to a higher SOC, thus providing scope for a further improvement in electrolyte utilization. The model calculates the cell or power module voltage during discharge at constant power. The ratio of the voltage under load to the open circuit voltage (OCV) gives the electrochemical efficiency at a given SOC, from which the average electrochemical efficiency across the SOC range investigated is computed.

The net power required per cell, P_{req} , is related to the power produced by the cell, P_{cell} , by Eq. 1:

$$P_{req} = P_{cell} - P_{pump} - P_{shunt} \quad [1]$$

Where P_{cell} is the actual cell output power, P_{pump} is the pumping power loss per cell and P_{shunt} is the power loss due to shunt current. This can be treated as a quadratic equation with respect to cell area, as the pumping losses increase with the square of the area, while the power produced is directly proportional to the cell area. After the cell area is determined to solve this equation, the total current is calculated from the input current density, and the flow rate per cell calculated from the input flow rate per current.

The next step is to calculate the effective voltage V_{eff} at each SOC from the cell voltage V , using Eq. [2].

$$V_{eff} = V \left(\frac{P_{cell} - P_{pump} - P_{shunt}}{P_{cell}} \right) \quad [2]$$

When the SOC is increased, the cell produces too much power at the same current density. Therefore, the current density is reduced iteratively using the Newton Raphson method, also referred to as Newton's method [37], until the system produces the correct amount of power (Eq. 1). Lower current density increases V_{eff} as the SOC increases, thus minimizing the energy-related costs by decreasing the amount of electrolyte needed. Each time the current density is

changed, the flow rate per cell, Q , is also changed to maximize V_{eff} . This is also done via Newton Rhapson iterations, with $Q_{i+1} = Q_i - \left(\frac{dV_{eff}}{dQ} \right) / \left(\frac{d^2V_{eff}}{dQ^2} \right)$ until V_{eff} is maximized, where i is the iteration step.

This is done across the entire SOC range to determine an average V_{eff} , $\overline{V_{eff}}$, which along with the ampere-hour capacity of the electrolyte per unit volume (Ah L⁻¹) and the depth of discharge (DOD), determines the effective electrolyte energy density, Wh_{eff} L⁻¹. The required electrolyte volume is calculated from the total energy required for the application.

With the cost C_s calculated at various operating conditions and stack designs that provide the required power and energy, additional Newton Rhapson iterations are performed to optimize the cost. The following steps are iterated until the cost reaches a minimum and stops changing more than 0.001 \$ kWh⁻¹ as shown in Eq. 3:

$$j_{i+1} = j_i - \left(\frac{dC_s}{dj} \right) / \left(\frac{d^2C_s}{dj^2} \right) \quad [3a]$$

$$q_{i+1} = q_i - \left(\frac{dC_s}{dq} \right) / \left(\frac{d^2C_s}{dq^2} \right) \quad [3b]$$

$$DOD_{i+1} = DOD_i - \left(\frac{dC_s}{dD} \right) / \left(\frac{d^2C_s}{dD^2} \right) \quad [3c]$$

Where j is the current density, q is the flow rate per unit current per cell, and DOD is the depth of discharge.

The program, as written, lends itself to exploring optimization with respect to additional parameters, such as the aspect ratio for each cell, defined as the ratio of cell width to height, and the flow channel dimensions.

The stack area corresponding to minimum total system capital costs varies with the specific energy and power requirements of the grid service being offered. While operating the stack at a higher power density results in lower stack area or lower power costs, it decreases the electrochemical efficiency, resulting in higher energy costs. Since power module costs are expected to dominate system costs at high Power to Energy ratio, running the stack at high power density is expected to provide the optimum solution. The reverse is expected to be the case for grid services that are energy intensive. During the design stage, flow battery manufacturers can use this tool to optimize the power module size for the various grid services these battery systems are being developed to offer.

By choosing the best operating conditions for each chemistry, various chemistries were compared on an “apples to apples” basis. All vanadium, iron vanadium and iron chromium chemistries were analyzed with this methodology.

Based on operating life, cost of capital, fixed charge rate, capital recover factor and other economic parameters, levelized cost was determined for public and private utilities [20].

Results

For all the chemistries, the analysis was carried out for E/P ratios of 0.25 and 4 to reflect the range of application duty cycles the storage systems are expected to be subjected to. For select cases, cost sensitivity analysis was done across an E/P range of 0.1 to 40.

Loss analysis

Various losses as a percentage of system power output across the SOC range for the optimum case are shown in Fig. 2 for conventional flow for V-V and Fe-V systems. The electrochemical loss is defined as the OCV divided by the actual voltage, while the shunt current and pumping losses are defined as the power loss divided by the total cell power output without

including the shunt current and pumping losses. As expected, the higher current density and lower concentration at low SOC conditions required the highest flow rate, with the associated largest pressure drop across the SOC range. For the V-V 4 hour case, the pumping losses range from 8% at low SOC to 2% at high SOC. Shunt current losses are highest at the high SOC due to high cell voltage, with a maximum loss of 1.4%. As expected, the lower concentration of Fe-V chemistry coupled with lower cell voltage corresponds to greater flow rate related losses across the entire SOC range, while shunt current related losses are lower across the SOC range due to lower cell voltage. For the sake of brevity, similar figures for the 0.25-h operation are not shown.

Table 2 shows the various losses for V-V and Fe-V systems. Electrolyte costs drive system costs for energy intensive applications. Operating the stack at low power density for high E/P ratios minimizes system costs, since the associated lower electrochemical losses improve electrolyte utilization. On the other hand, at low E/P, while running the stack at higher power density increases electrochemical loss, the system cost decreases due to low stack component costs. The V-V system has lower flow rate related losses than Fe-V due to its higher active species concentration, lower viscosity and higher cell voltage. The lower Fe-V cell voltage contributes to smaller current shunting losses.

The pumping losses are higher for the 0.25-h Fe-V than for the 4-h case due to a higher optimum current density requiring a higher flow rate. Contrary to expectations, the pumping loss as a percentage of total battery system output is slightly higher for V-V 4h than V-V 0.25h. While the optimum current density is the same for both cases, the higher SOC range associated with the 4-hour operation requires higher flow rate to provide the required power at low SOC. Note that the average pumping losses are 30 kW and 26 kW for the V-V 4-h and 0.25-h scenarios respectively. Since the power costs are dominating for the V-V 0.25-h case, sacrificing

electrochemical efficiency by using lower flow rate at low SOC provides the most cost optimum solution. Higher flow rates lead to an increased pump cost that exceeds the decrease in stack and energy costs related to improved performance. Identification of such subtleties can be immensely helpful during stack design, development and operation for the vast number of grid services and their diversity in terms of storage duty cycles.

While Table 2 provides a summary of total losses for each chemistry, this table, in conjunction with Fig. 2 forms the basis for the development of a battery management system that can be used by system developers and end users.

Cost analysis

Capital cost calculations were performed by optimizing the operating parameters for the lowest cost. The total cost in \$ kWh⁻¹ is shown for an optimized 1 MW conventional flow system with energy requirements of 0.25 MWh (Fig. 3a) and 4 MWh (Fig. 3b). This optimization algorithm minimizes the chemical costs by increasing the SOC range and electrochemical efficiency for the 4-h case, and minimizes the stack costs by decreasing the SOC range and increasing the current density for the 0.25-h case.

The power conversion system (PCS) cost for the 1 MW system is the same across all chemistries, and is 46 \$/kWh for E/P=4 and 735 \$/kWh for E/P=0.25. . For the 0.25-h case, since PCS costs are a high fraction of total costs, they have been shown as a separate category in Fig. 3a, while for the 4-h case, the PCS cost has been grouped with pumps, frames, bolts into the “other” category.

V-V has the lowest cost for both energy to power ratios due to its high cell voltage, high concentration and low viscosity. Fe-V is more cost effective than Fe-Cr for the power intensive case due to higher maximum current density, with the reverse true for the energy intensive case

due to lower costs for Fe-Cr electrolyte. Chemical costs tend to dominate for the 4-h case, while stack components costs (bipolar plates, felt electrodes and membrane) for the 0.25-h case. The total costs are broken down into component costs to explore where improvements can be made in redox flow battery development.

For the V-V battery system, the membrane costs contribute 18% of total cost of 2080 \$ kWh⁻¹ at E/P = 0.25, while chemical costs are at 14%. Note that since PCS costs are 35% of total system costs, the membrane and chemicals contribute 27% and 22% respectively on a PCS-free basis. Hence, improving power density is expected to minimize system cost. This can be achieved by higher electrolyte conductivity, improved electrode kinetics, and electrodes with higher active specific area. Note that the optimum operating point for a power intensive application does not necessarily correspond to the higher energy efficiency. For E/P = 4, chemicals cost dominate at 58% of total cost of 325 \$ kWh⁻¹. Hence, the optimization algorithm lowers costs for high energy battery systems by increasing stack efficiency and SOC range by operating at lower power density (as seen in Table 3) compared to power intensive grid services to reduce chemical costs at the expense of increased stack or power module costs.

For the Fe-V system, bipolar plates at 12% and felt electrode at 25% combine for 37% of system **costs at** E/P of 0.25. Chemical costs at 630 \$ kWh⁻¹ are more than double the chemical cost for the V-V system at \$290/kWh, comprising 18% of total costs. Since PCS costs are 21% of total costs, the bipolar plates and felt electrodes contribute 47% of PCS-free total system costs, while chemical costs comprise 23% of PCS-free total system costs. For the 4-h application, chemical costs at 360 \$ kWh⁻¹ (60% of total costs) is 1.9 times greater than the V-V 4-h system chemical costs at 185 \$ kWh⁻¹, nearly the same ratio as the total system costs for the two chemistries. As expected, the Fe-V chemical costs are higher due to its lower cell voltage.

For the Fe-Cr system, the power module components dominate at 79% for the 0.25-h case, with felt and bipolar plate costs of 41% and 17% respectively. However, even for E/P of 4, the low maximum current density of 50 mA cm^{-2} leads to high power module component costs of 62%, with felt and bipolar plate costs at 32% and 13%, respectively, in addition to high chemical costs of 26%.

Table 3 shows unit power, energy and total costs per unit energy, along with the optimized conditions to operate the V-V and Fe-V systems. Even though the Fe-V system uses a significantly cheaper separator than the V-V system, the power costs for V-V are still lower due to the much smaller stack size associated with the V-V system's higher power density. The electrode areas for the V-V system are 1490 cm^2 and 1850 cm^2 for E/P ratios of 0.25 and 4 respectively, with the corresponding electrode areas for the Fe-V system equal to 7120 cm^2 and 10900 cm^2 . The low cell voltage of Fe-V also leads to a much higher unit energy cost. Note that for the V-V system, the model predicts a higher optimum current than the value of 320 mA cm^{-2} shown in Table 3. However, since this is the highest experimentally verified current density in our laboratory, the current density is capped at 320 mA cm^{-2} . Both chemistries had a lower optimum SOC range for the 15-minute operation, as this minimizes cell area and pump size, while a larger SOC range for the 4-h operation minimizes electrolyte cost. The flow rate normalized per unit current at low SOC was lower for the 0.25-h case to minimize power related costs. The lower depth of discharge (DOD) or higher minimum SOC for the 0.25-h case also contributes to the lower normalized flow rate, since ΔSOC effects are not significant for a shallow DOD. For both the E/P ratios, the current-normalized flow rate was lower for the Fe-V system to keep pressure drop losses and pump costs low, since this chemistry requires a higher total current due to its lower voltage. If the number of cells/stack or the number of stacks were to

be increased to keep the total DC voltage nearly the same as a V-V system, the Fe-V system would still require a higher flow rate for the same current-normalized flow rate, since more number of cells are needed to provide the required power at a fixed total current. As discussed earlier, the optimum power density for both chemistries is lower for $E/P = 4$, with V-V power density about 6 times higher than that for the Fe-V system.

A cost sensitivity analysis determined the effect of the maximum current density, which as discussed earlier, occurs at low SOC. For the $\frac{1}{4}$ -h operation, the vanadium chemistry was most sensitive to current density, with system costs decreasing by 33% from 3100 \$ kWh⁻¹ when the current density is increased from 100 to 320 mA cm⁻² (Fig. 4a). For the same current density range, the 4-h all vanadium system cost decreased by only 14%. As stated earlier, 320 mA cm⁻² was the highest current density obtained in our laboratory. While the cost for the 4-h application appears to have plateaued at 300 mA cm⁻², there is significant room for cost reduction by moving to even higher current density for the $\frac{1}{4}$ -h case. In the narrow current density range investigated for the Fe-Cr system, the cost decreased with increasing current density for both E/P ratios. It is anticipated that as more data become available for this system, the current density would extend to higher values, with associated decrease in costs. For the Fe-V system, the cost minimum occurred at 155 mA cm⁻² for E/P of 0.25, with a broad minimum in the 100-125 mA cm⁻² range for E/P of 4. In other words, for the Fe-V chemistry, high current densities, accompanied by lower electrochemical efficiency, drive chemical costs up to adversely affect total system cost even at low E/P ratios. The effect of lower electrochemical efficiency on high Fe-V chemical cost is also seen in Fig. 3a, where the chemical costs contribution for the Fe-V chemistry is more than two times higher than that for the V-V chemistry at E/P of 0.25.

At 320 mA cm^{-2} where V-V costs are minimized, membrane and chemicals were the most important contributors to total cost. Using baseline cost input values, varying the membrane unit costs within the range investigated in this study resulted in system costs of 1860 to 2080 $\text{\$kWh}^{-1}$, while the corresponding range was 1890 to 2080 $\text{\$kWh}^{-1}$ by varying chemical costs for the 0.25-h case (Fig. 5a). Clearly, electrochemical efficiency is still a driver of system costs at low E/P, as reflected in the sensitivity to chemical costs. The sensitivity analysis for the 4-h service is shown in Fig. 5b. The system cost is highly sensitive to chemicals, decreasing by 38% from the baseline value of 325 $\text{\$ kWh}^{-1}$, while varying the membrane cost had a much smaller effect, with the cost decreasing by a mere 3% from 325 $\text{\$ kWh}^{-1}$.

The system cost is significantly impacted by chemical costs, maximum current density and membrane costs at low E/P ratio. For high E/P ratio, with chemical costs dominating, improving electrolyte utilization and system efficiency is expected to decrease costs. This can be achieved by increasing the SOC range of operation and improving system efficiency through better kinetics, higher electrolyte conductivity and lower pumping losses using better flow path design such as interdigitated flow.

Fig. 5c presents the total Fe-V system cost as a function of chemicals and felt electrodes costs for the ¼ h system at the most cost-effective current density 152 mA cm^{-2} . Lowering felt electrode costs decreased unit energy cost from $3460 \text{ \$ kWh}^{-1}$ by 18%, while the corresponding value was 12% for lower chemical costs. For the 4-h case (Fig. 5d), at an optimum 111 mA cm^{-2} , V_2O_5 was a significant cost driver, the cost dropping from 595 kWh^{-1} to 355 kWh^{-1} as V_2O_5 price drops from 24 kg^{-1} to 8 kg^{-1} . As felt cost was lowered, the cost decrease of 60 kWh^{-1} was much lower, showing felt cost is not the driver of system cost at high E/P ratio.

The system cost at various E/P ratios for all the chemistries were plotted as shown in Fig. 7, for both present (Fig. 6a) and optimistic scenarios (Fig. 6b) to demonstrate the significance of various grid services duty cycles profiles on system costs. The corresponding normalized cost using V-V system cost as basis is given in Fig. 6c and 6d. For the Fe-Cr chemistry, data from Figure A-1 of reference [27] was used for performance curves for the optimistic case, since a higher maximum current density of 75 mAcm^{-2} was used in this work. With the availability of performance curves from the Fe-Cr system developers, it is expected that higher current densities are achievable, which could lower costs further for the Fe-Cr system.

The Fe-Cr system is cheapest at E/P ratio > 10 for the present case (Fig. 6a and 6c), with the cost $230 \text{ \$ kWh}^{-1}$ for both V-V and Fe-Cr at 10 hours, while Fe-Cr cost, at $410 \text{ \$ kWh}^{-1}$, is 27% higher than all vanadium cost for the 4-hour application. At high E/P ratio, the V-V costs are higher than Fe-Cr system costs for the present case, since the system costs are dominated by chemicals. Since chemicals and membrane costs are a major percentage of the V-V system present costs, the anticipated drop in chemicals and membrane cost in the optimistic scenario enables the V-V system cost to be slightly lower than Fe-Cr even at $\text{E/P} > 10$ (Fig. 6b and 6d). However, with availability of latest Fe-Cr performance curves, estimated costs are expected to

decrease for this system also. Hence it is too early to draw definitive conclusions regarding these systems at high E/P ratios.

For the present costs, Fe-V is more cost effective than Fe-Cr system costs at E/P ratio < 1. For the optimistic scenario, the improved performance curves used for Fe-Cr **are** expected to remove the cost advantage of Fe-V systems at low E/P ratio, with the costs being equal at E/P of 0.1. At high E/P ratio, the Fe-V system is the most expensive, due to chemical costs dominating total system costs.

In the optimistic scenario, all vanadium system costs are 165 \$ kWh⁻¹ for a 4-h operation, while Fe-Cr costs are 10% higher. The 10-h costs are 105 \$ kWh⁻¹ and 110 \$ kWh⁻¹ for V-V and Fe-Cr respectively. These numbers indicate that flow battery energy storage is indeed expected to be cost effective as component costs decrease and performance improves.

A user-friendly tool developed for this model has been released with open access to enable utilities, battery manufacturers and other stakeholders to test the effect of various operating parameters on stack sizing and cost [29] to enable design of cost effective battery systems for multiple grid services. Several stakeholders have used this tool and provided feedback.

Interdigitated Flow Analysis

Interdigitated cells can be used to reduce pressure drop within the felt. Flow channels are cut into the felt using a laser (PNNL unpublished work), resulting in a shorter length traveled by the electrolyte, and therefore a lower pressure drop. Schematics of flow through conventional flow and interdigitated flow are presented in Figs. 12 and 13 of [21]. Systems with interdigitated flow path were modelled to investigate the effect on cost. The channels run along the height of the cell, while electrolyte flows through the electrodes parallel to the width of the cell, with the

height and width equal to each other. Hence, in terms of the pressure drop calculation within the felt electrode, the flow path length is simply the inter-channel length, while the width is the entire height of the electrode. This leads to a very high aspect ratio, defined as width to flow path length. As seen from Eq. 1, this lowers the pressure drop considerably. Based on mechanical considerations, the interdigitated flow channels are assumed to be a minimum of 10 cm apart.

V-V and Fe-V systems were modelled using interdigitated flow at various E/P ratios, with results for E/P ratio of 0.25 and 4 shown in Table 4. Interdigitated flow path decreased system costs for both the chemistries and both E/P ratios, with the decrease being greater at E/P of 4.

For the 0.25 h case, the use of interdigitated flow fields decreased cost by 12% for Fe-V and 1% for V-V systems. For the 4 h case, the corresponding cost reductions were 14% and 6%.y. The table also provides costs for the near term and optimistic scenarios. As component costs decrease, the effect of interdigitated flow is less, as expected. For the optimistic scenario, a 50 -55% drop in cost compared to the present costs is seen for the 4-h application, with all vanadium chemistry costs projected at 160 \$ kWh⁻¹ for the interdigitated flow pattern, while the cost is nearly the same for conventional flow.

The Fe-V system benefits more from interdigitated flow path for both E/P ratios. At an E/P ratio of 0.25, for the Fe-V system, the optimum current density is higher by 24%, enabled by an increase in maximum flow rate of 46%, with an expansion of the SOC range by 13%, while the corresponding numbers for the all vanadium system are 0%, 6% and 3%. The interdigitated flow for the 0.25-h application benefits the Fe-V by increasing the current or power density, thus decreasing power component costs, and by increasing the SOC range, lowering energy related costs. For the V-V chemistry, with the current density capped at 320 mA cm⁻², the flow rate for

interdigitated design is limited by incremental pump costs that exceed the cost savings from reduced pressure drop losses and higher electrochemical efficiency. As seen in Fig. 4a, for the 0.25-h case, there is room for cost decrease for the V-V system by increasing the current density further, in which case interdigitated flow is expected to provide more cost savings.

For the E/P ratio of 4, for the Fe-V system, the optimum current density is higher by 27%, accompanied by a 59% increase in the maximum flow rate and an increase in the SOC range by 11%, thus decreasing power and energy related costs. The corresponding numbers are 0%, 20% and 4% respectively for the all vanadium system. The low cell voltage for Fe-V lends itself to being more suited for interdigitated flow, since expansion of the allowable SOC range is critical to lowering system costs. In addition, since the volumetric energy density of the Fe-V is lower than the all vanadium system, the higher flow rates at the same stoichiometry for this system also makes interdigitated flow a suitable choice for this chemistry. This concept can be used for other novel flow battery systems with low cell voltage. For the V-V system, interdigitated flow, while not as effective for the Fe-V chemistry, does lower costs by 6% by expansion of the SOC range and improvement of electrochemical efficiency enabled by a 20% increase in flow rate. This effect will be enhanced further for applications with a higher E/P ratio such as renewable time shifting, outage mitigation and arbitrage.

Model Validation

To validate the model, the estimated system cost was compared against results obtained using experimental data for multi-cell V-V redox stack using cells with an active electrode area of 26 cm by 30 cm (780 cm^2) [38]. This was performed for 15-cell stack, for both normal flow and flow through interdigitated channels (ID), and for a 3-cell stack with interdigitated flow. The stack was run at various current densities and various flow rates, using constant current and flow rate throughout discharge.

The battery performance at various operating conditions was used to compute system cost. The cost was calculated for E/P ratio of 4 for two cases:

- (i) The cost per unit power and energy were calculated from stack data for the stack with electrode area of 780 cm^2 , from which $\$ \text{ kWh}^{-1}$ was calculated for E/P of 4.
- (ii) The stack voltage curves were used to determine power density, and the required cell area at low SOC calculated for a 1 MW/4 MWh system consisting of 36 60-cell stacks.

The system cost was then computed as described earlier.

Comparison of the above costs was done using the electrochemical model to generate stack performance curves under the same operating conditions (SOC range, current density and initial flow rate) for cases i and ii. The operating parameters were subsequently optimized at constant current density to provide the minimum system costs. The optimization was repeated in the constant power mode to further reduce costs by lowering pumping losses, since the stack operates at lower current density at all SOC's greater than the low end of the SOC range.

Fig. 7 presents the results for both cases, with Fig. 7a corresponding to the fixed 780 cm^2 electrode area (case i) and Fig. 7b corresponding to the cell area required for a 1 MW/4 MWh system (case ii). For interdigitated flow, unit energy cost from the model was on average 2.5%

lower than the cost from stack data (Fig. 7a). Using optimized operating conditions brings the cost down 12%, while operating at constant power reduces the cost by an additional 2%.

For conventional flow, the electrochemical model cost for the 15-cell stack was 3.5% higher than the calculated cost from stack data. Note that the $2.1 \text{ ml min}^{-1} \text{ A}^{-1}$ flow rate for conventional flow was too low, causing the model to be unstable at low SOC. Hence, the SOC was restricted to 31-69% to compare the stack data costs with the model estimate, resulting in a 25% and 32% increase in system costs compared to the interdigitated flow for the stack data and model estimate respectively. Using optimized operating conditions brings the cost down 32%, while operating at constant power reduces the cost by an additional 3%.

The estimated costs for the 1 MW 4 MWh system are given in Fig 7b. For the interdigitated cases, the cost for the electrochemical model is 1-3% lower than the cost obtained from stack data. Optimized operating conditions bring the cost down 10%, while operating at constant power further reduces costs by 2%. For conventional flow, the electrochemical model cost is 1% lower than the cost from stack data. Due to the restricted SOC range as discussed earlier, the costs are 34% higher than corresponding costs for the interdigitated flow. Using optimized operating conditions brings the cost down 34%, while operating at constant power reduces the cost by an additional 3%.

From both Figs. 7a and 7b, the trends are very similar for both the fixed cell area of 780 cm^2 (case i) and 1 MW (case ii) scenarios, showing the scalability of the model.

The current density, flow rate, SOC range and total system cost for the 15-cell interdigitated, 780 cm^2 stack are given in Table 5, while corresponding values for conventional flow are given in Table 6. For interdigitated flow (Table 5), the optimized cost for the electrochemical model is 12% lower than the non-optimized model cost of $\$470 \text{ kW}^{-1}$. The main

reason is the expansion of the SOC range from 21-79% to 11-89%, coupled with an increase in current density to 305 mAcm^{-2} , driving down both energy and power related costs. The approximate doubling of the current-normalized flow rate to $8.4 \text{ ml min}^{-1} \text{ A}^{-1}$ at low SOC does result in increased pump costs, while pumping losses are also increased due to higher flow rates across the entire SOC range. However, as discussed earlier, for energy intensive applications, lowering the energy cost is paramount. The higher flow rate allows for better electrochemical performance, enabling a much wider SOC range of operation. For operation at constant power, the cost drops by an additional 2% compared to the optimized constant current run, due to use of lower current density at high SOC, accompanied by lower flow rate/pressure drop losses and higher electrochemical efficiency.

Table 6 shows the corresponding numbers for conventional flow for the 15-cell 780 cm^2 stack. The optimized cost for the model is 34% lower than the non-optimized case, due to the very low current-normalized flow rate of $2.1 \text{ ml min}^{-1} \text{ A}^{-1}$ used for this run.. The optimized flow rate per cell per amp is about 3.5 times the non-optimized flow rate at low SOC, and twice at the high end of SOC (not shown in the table), thus demonstrating the importance of flow rate in driving overall system performance and cost. Operation at constant power further lowers the cost by 3%, mainly due to lower current density at higher SOC leading to better electrochemical efficiency, with the associated lower flow rate resulting in lower pumping losses.

For this chemistry and electrode design, the optimized cost for interdigitated flow pattern is 2.5% lower than that for conventional flow at both constant current density and constant power. Work is ongoing to determine the effective electrode thickness in each electrode, and determining appropriate felt electrode design for each polarity as a function of E/P ratios. We expect the advantages of interdigitated flow fields to be significantly higher using these considerations.

The system cost for V-V and Fe-V in the E/P range of 0.1-40 is presented in Fig. 8 for interdigitated flow. Just as for conventional flow, the V-V chemistry is more cost effective than Fe-V for the entire E/P range. For a 10-h application, the V-V cost is 220 \$ kWh⁻¹, while Fe-V is 375 \$ kWh⁻¹. The numbers for a 40-h service are 165 \$ kWh⁻¹ and 285 \$ kWh⁻¹ respectively, thus demonstrating the feasibility of the flow battery system across a wide range of applications.

The drop in cost for interdigitated flow over conventional flow increases with increasing E/P ratio for both chemistries, due to better utilization (higher SOC range) of the electrolyte. The Fe-V drop in costs is in the 10 to 18% range, while the corresponding range for the V-V system is 1 to 8%. As discussed earlier, interdigitated flow benefits the iron vanadium chemistry more across the entire E/P range, by reducing both power and energy costs. Once current density higher than 320 mA cm⁻² is demonstrated for the V-V system, interdigitated flow is expected to further lower costs further.

Table 7 presents the results for the levelized costs for the all vanadium and iron vanadium system for 4-h case with interdigitated flow using present costs. The all vanadium levelized cost for public utilities is 16 cents kWh⁻¹, 22% lower than that for investor owned utilities. The levelized cost for Fe-V is 30 cents/kWh for private utilities, while the public utility costs is 25% lower at 22 cents/kWh. The all vanadium levelized costs is around 30% lower than Fe-V

levelized costs, and is expected to be competitive with conventional generation with further drop in component costs and improvements in performance.

Conclusions

The redox flow battery systems model optimizes operation parameters to minimize costs, providing valuable insights into the contribution of various losses for different chemistries across a wide range of energy to power ratios. Model validation was done for in-house 3-cell and 15-cell stack data for a 2 kW and 1 MW system, demonstrating the scalability of this model. Using our validated tool, it has been demonstrated that an optimized all-vanadium system has an estimated system cost of $< \$350\text{kWh}^{-1}$ for an energy to power ratio of 4. With anticipated decrease in component costs coupled with improved performance, the system cost is expected to decrease to 160 kWh^{-1} for a 4-h application, and to $\$100\text{ kWh}^{-1}$ for a 10-hour application.

Chemistries most suited for various applications were identified. The component cost distribution and cost sensitivity to various components for a range of energy to power ratios guide future R&D efforts. For the all vanadium system, interdigitated flow was 6% more cost effective than conventional flow for an energy to power ratio of 4, with further cost reductions possible at higher E/P ratio. For the low voltage Fe-V systems, interdigitated flow effects are significantly higher across all E/P ratios, with a 12% and 14% decrease in cost for the 0.25-h and 4-h applications respectively. Interdigitated flow supports a 25% higher current density and expansion of the SOC range of operation enabled by 50% higher flow rates.

This work enables the redox flow battery community to design systems that minimize costs for various grid services. A systematic approach to continuously adjust battery operating conditions such as current density and flow rate with varying battery SOC, while also selecting

the DOD for various applications is provided. A user-friendly tool has been released for the industry stakeholders to test drive this model for their specific chemistry and preferred grid service.

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Table Captions

Table 1. List of symbols

Table 2. Summary of all losses for various chemistries and energy/power ratios for conventional flow

Table 3. Unit power and energy costs and total cost per kWh for 1 MW, 4 MWh system for conventional flow

Table 4. Comparison of results for interdigitated vs. conventional (normal) flow

Table 5. Model validation based on V-V stack data for 15 cell, 780 cm² stack with interdigitated (ID) flow pattern

Table 6. Model validation based on V-V stack data for 15 cell, 780 cm² stack conventional flow pattern

Table 7. Levelized costs of electricity for 1 MW/4MWh interdigitated V-V and Fe-V flow system using present costs.

Figure Captions

Fig. 1. Flowchart of process for optimizing cost

Fig. 2. Estimated losses from the model for conventional flow in an optimized 1 MW, 4 MWh system for (a) V-V system and (b) Fe-V system

Fig. 3. Component and system costs for a 1 MW system for all chemistries; (a) 0.25 MWh and b) 4 MWh

Fig. 4. Effect of current density for 1 MW system for all chemistries for conventional flow (a) 0.25 MWh and b) 4 MWh

Fig. 5. Effect of dominant component costs for a) V-V 1 MW, 0.25 MWh, b) V-V 1 MW, 4 MWh, c) Fe-V 1 MW, 0.25 MWh, d) Fe-V 1 MW, 4 MWh

Fig. 6. . Effect of E/P ratio on cost of 1 MW system for a) Present case and b) Optimistic case, with normalized costs (ratio of cost to V-V cost) for c) Present case and d) Optimistic case.

Fig. 7. All vanadium stack performance data and electrochemical model performance for a) 15 cell conventional flow, b) 15 cell ID, and c) 3 cell ID

Fig. 8. Total system cost at various E/P ratios for interdigitated (ID) flow (solid lines) and % drop in cost over conventional flow (dotted lines).

Table 1. List of symbols

Symbol	Description
C	Molar concentration
C_3	Cost Function
D_H	Hydraulic Diameter
DOD	Depth of discharge
F	Faraday's constant
h	Cell height (direction of flow)
I	Current
j	Current density
K	Permeability
l	Length of flow channel
ΔP	Pressure drop
P	Power
Q	Flow rate per cell
q	Flow rate per current per cell
t_{cell}	Cell thickness
V	Voltage
w	Cell width (perpendicular to flow)
η	Pumping efficiency
μ	Dynamic viscosity

Table 2. Summary of all losses for various chemistries and energy/power ratios for conventional flow.

	Electrochemical	Pump	Shunt	Total
E/P = 0.25h				
V-V Gen 2	17%	2.0%	1.0%	20%
Fe-V	26%	6.2%	0.2%	32%
E/P = 4h				
V-V Gen 2	15%	2.3%	1.1%	18%
Fe-V	18%	5.1%	0.2%	23%

Table 3. Unit power and energy costs and total cost per kWh for 1 MW, 4 MWh system for conventional flow.

Chemistry	\$ kW ⁻¹	\$ kWh ⁻¹	Total \$ kWh ⁻¹	SOC Range %	Current density mA cm ⁻²	Max flow rate ml min ⁻¹ A ⁻¹	Cell Area cm ²	Power density W cm ⁻²
E/P = 1/4								
V-V Gen 2	445	290	2080	24-76	320	3.6	1490	0.31
Fe-V	705	630	3460	26-74	155	3.1	7120	0.07
E/P = 4								
V-V Gen 2	535	190	325	11-89	320	6.9	1850	0.25
Fe-V	940	360	595	13-87	110	3.9	10920	0.04

Table 4. Comparison of results for interdigitated vs. conventional (normal) flow

Scenario	Total \$ kWh ⁻¹			SOC Range %	Current mA cm ⁻²	Max flow rate ml min ⁻¹ A ⁻¹
	Present	Near Term	Optimistic			
E/P = 0.25h						
Fe-V Normal	3460	2680	1940	26-74	155	3.1
Fe-V ID	3060	2450	1870	23-77	195	4.5
V-V Normal	2080	1730	1370	24-76	320	3.6
V-V ID	2050	1720	1370	23-77	320	3.8
E/P = 4h						
Fe-V Normal	595	430	265	13-87	110	3.9
Fe-V ID	510	375	240	9-91	140	6.1
V-V Normal	325	245	165	11-89	320	6.9
V-V ID	305	235	160	9-91	320	8.3

Table 5. Model validation based on V-V stack data for 15 cell, 780 cm² stack with laser-cut interdigitated (LID) flow pattern

Scenario	Total \$ kWh ⁻¹	SOC Range %	Current mA cm ⁻²	Flow rate ml min ⁻¹ A ⁻¹
E/P = 4h				
Stack Data 15 Cell LID	480	21-79	240	4.3 (constant)
EC Model Same Parameters	470	21-79	240	4.3 (constant)
EC Model Optimized Parameters	410	11-89	305	8.4 (Maximum)
EC Model Optimized Parameters Constant Power	400	10-90	320	8.3 (Maximum)

Table 6. Model validation based on V-V stack data for 15 cell, 780 cm² stack conventional flow pattern

Scenario	Total \$ kWh ⁻¹	SOC Range %	Current mA cm ⁻²	Flow rate ml min ⁻¹ A ⁻¹
E/P = 4h				
Stack Data 15 Cell Normal	600	31-69	240	2.1 (constant)
EC Model Same Parameters	620	31-69	240	2.1 (constant)
EC Model Optimized Parameters	420	11-89	300	7.8 (Maximum)
EC Model Optimized Parameters Constant Power	410	10-90	320	7.5 (Maximum)

Table 7. Levelized costs of electricity for 1 MW/4MWh interdigitated V-V and Fe-V flow system using present costs.

	\$ kWh ⁻¹	
	Private	Public
V-V	0.21	0.16
Fe-V	0.30	0.22

Figure(s)

Fig. 1. Flowchart of process for optimizing cost

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(a)

(b)

(c)

(d)

Fig. 5. Effect of dominant component costs for a) V-V 1 MW, 0.25 MWh, b) V-V 1 MW, 4 MWh, c) Fe-V 1 MW, 0.25 MWh, d) Fe-V 1 MW, 4 MWh

(a)

(b)

(c)

(d)

Fig. 6. Effect of E/P ratio on cost of 1 MW system for a) Present case and b) Optimistic case, with normalized costs (ratio of cost to V-V cost) for c) Present case and d) Optimistic case

Fig. 7. Validation of electrochemical performance model for $E/P = 4$ using V-V stack data a) 780 cm^2 fixed electrode area b) varying electrode area to get 1 MW

Fig. 8. Total system cost at various E/P ratios for interdigitated (ID) flow (solid lines) and % drop in cost over conventional flow (dotted lines).