Contents lists available at ScienceDirect





Electrochimica Acta

journal homepage: www.journals.elsevier.com/electrochimica-acta

Capital cost evaluation of conventional and emerging redox flow batteries for grid storage applications



L. Tang^a, P. Leung^a,^{*}, M.R. Mohamed^b, Q. Xu^c,^{*}, S. Dai^a, X. Zhu^a, C. Flox^d,^{*}, A.A. Shah^a, Q. Liao^a

^a Key Laboratory of Low-grade Energy Utilization Technologies and Systems, MOE, Chongqing University, Chongqing 400030, China

^b Sustainable Energy & Power Electronics Research Group, Faculty of Electrical & Electronics Engineering, University Malaysia Pahang, Pekan, Pahang 26600, Malaysia

^c Institute for Energy Research, Jiangsu University, 301 Xuefu Road, Zhenjiang 212013, China

^d Institut de Ciencia de Materials de Barcelona, CSIC, Campus UAB, Barcelona 08193, Spain

ARTICLE INFO

Keywords: Aqueous Capital cost Non-aqueous Redox flow batteries Techno-economic analysis

ABSTRACT

Redox flow battery (RFB) is a promising technology to store large amounts of energies in liquid electrolytes attributable to their unique architectures. In recent years, various new chemistries have been introduced in both aqueous and non-aqueous electrolytes as pathways to lower-cost systems, eventually meeting the long-term cost target of USD\$ < 100 (kW h)⁻¹ for board market penetration. Since there is a lack of capital cost data available for flow batteries under the same criteria and assumptions, a fact-based techno-economic analysis is evaluated based on real systems to facilitate the explorations of more competitive systems. In total, nine conventional and emerging flow battery systems are evaluated based on aqueous and non-aqueous electrolytes using existing architectures. This analysis is attempted to evaluate the feasibility of these emerging systems to meet the cost target and to predict their technological prospects for energy storage applications. The capital costs of these resulting flow batteries are compared and discussed, providing suggestions for further improvements to meet the ambitious cost target in long-term.

1. Introduction

For modern electrical grids, large-scale energy storage systems are often coupled with conventional and renewable power generations to provide stable outputs for various applications ranging from uninterruptible power to load-levelling [1,2]. Rapid growth of these systems has received significant attentions from both academia and industries. It is important to store excess electricity generated from conventional power plants and intermittent renewable energy sources grid-connected and off-grid. Pumped hydro storage is still the commonest and the most cost-effective form of energy storage, accounting for more than 95 % of installed global capacities [3]. However, due to the concerns of environmental issues and the dependency of specific terrains, it remains challenging to find suitable sites for implementing this type of energy storage [4].

By comparison, electrochemical energy storage systems, such as batteries, have the advantages in terms of operational flexibilities and are not limited by geographical constraints. The use of batteries has increased rapidly due to their versatility and scalability for various applications, from consumer electronics to load-levelling. The global demand of batteries is expected to grow 25 % annually from 185 GW h in 2020 to over 2,000 GW h by 2030 [5]. For the United States and China, the demands of using batteries for energy storage and electrification of transport will increase by more than 100 and 10 times, respectively. By comparison, such demand for consumer electronics is expected to grow less than twice and will represent only < 5 % of the global battery market [6].

Despite the high energy densities, the performance of lithium-ion batteries degrades rapidly under over-charge or deep discharge conditions. Importantly, they are also considered not suitable for storing energies at large-scale (such as load-levelling) due to the increasing safety concerns in cases of failure/thermal events [7,8]. Unlike most batteries, redox flow batteries have unique architectures that store all or part of their energies in liquid electrolytes instead of electrodes within cells. Since other batteries have a fixed energy to power (E/P) ratio, the architecture of flow batteries enables energy and power to be decoupled, which can be adjusted with the amount of the electrolytes and the sizes of the total electrode areas, hence the power rating is based on the stack

* Corresponding authors. E-mail addresses: leungpuiki@hotmail.com (P. Leung), xuqian@ujs.edu.cn (Q. Xu), cflox@icmab.es (C. Flox).

https://doi.org/10.1016/j.electacta.2022.141460

Received 19 September 2022; Received in revised form 24 October 2022; Accepted 30 October 2022 Available online 31 October 2022

^{0013-4686/© 2022} The Authors. Published by Elsevier Ltd. This is an open access article under the CC BY-NC-ND license (http://creativecommons.org/licenses/by-nc-nd/4.0/).



Fig. 1. Schematics and charge-discharge profiles of the selected aqueous and non-aqueous flow batteries evaluated in this techno-economic analysis [26–34].

size or number. For the same power output, only more electrolytes are needed to increase the discharge capacities/durations. By contrast, the cost per kW h of lithium-ion batteries tends to flatten out beyond 4 h storage duration as more cells and essential components are required. Due to the modular configurations, redox flow batteries are more scalable and have longer lifespans than other batteries, making them more suitable for energy storage in the range of kW/ kW h to MW/ MW h [9].

Over the past decades, although various flow battery chemistries have been introduced in aqueous and non-aqueous electrolytes, only a few flow batteries (i.e. all-V, Zn-Br, Zn-Fe(CN)₆) based on aqueous electrolytes have been scaled up and commercialized at industrial scale (> kW) [10–12]. The cost of these systems (E/P ratio = 4 h) have been evaluated in a range of USD\$ 350 — 600 (kW h)⁻¹ by several US national laboratories [13] and compared with other major energy storage systems (electrochemical and physical systems). However, new systems of using inexpensive and earth-abundant active materials (inorganic species and tailored organic molecules) have been introduced in different configurations using aqueous and non-aqueous electrolytes as pathways to lower-cost systems [14,15]. Also, advances in electrode and membrane materials have the tendency to reduce cost to a further extent. Thus, for applications from load-levelling to electric vehicles, the costs of these batteries vary with chemistries, performances and cell components. Realistic improvements of these aspects identified by cost evaluations shall facilitate the designs of new systems and cost reductions of meeting the Department of Energy (DoE, US) target of USD\$ $< 100 \text{ (kW h)}^{-1}$ for board market penetration in long-term [16].

In general, cost reduction of aqueous batteries is known to be achieved by decreasing the active material costs, considering the costs of water and its salts are almost negligible (USD\$ 0.1 kg^{-1}). However, it is also influenced by the aforementioned factors. The objectives of new systems are to obtain high solubility and cell voltages simultaneously, although the stability windows are limited to < 2.0 V due to water electrolysis. By contrast, non-aqueous systems (including > 3.0 V hybrid system with lithium anodes) enable possibilities of electrochemical stability, multi-electron transfers and increased solubilities, especially when organic active materials are used [17]. The main challenges are the high cost and the resistances of these salts and solvents. Therefore, increasing power density is considered as the most effective strategy in cost reduction, since fewer cells and parts are required to deliver the same energy and power outputs.

Prior to this work, there is a lack of capital cost data available for flow batteries under the same criteria and assumptions, especially for those emerging or conceptual systems. Cost evaluations of a few aqueous flow batteries have been conducted using techno-economic analyses

[18,19]. There are also some investigations focusing on life-cycle assessments [20] and costs associated with the manufacturing of cell components [21], production/commercial pricing of active materials [22] and other miscellaneous items (i.e. depreciation, labour, overhead, profit and warranty for specific systems [20,21]. Conventional cost performance models were introduced by Sprenkle and co-workers based on electrochemical models taking account of pump losses and shunt current for 1 MW all-vanadium and iron-vanadium batteries [23]. These models were later expanded to other chemistries, e.g. iron-chromium, and showed the costs of these systems were still more than USD\$ 190 $(kW h)^{-1}$ for 8 h applications [24]. Since the durations or capacities are dependent on the electrolyte volumes/concentrations, the resulting electrolyte costs dominate with systems of longer durations/ smaller power ratings (as at high E/P ratios). Brushett and co-workers have developed several cost models to evaluate design pathways for aqueous and non-aqueous flow batteries by considering the material costs and performance parameters in the design iterations [25]. For instance, an ambitious cost target of USD $$100 (kW h)^{-1}$ can be achieved for aqueous systems with the combination of USD $\$ 2 kg⁻¹ active material, 100 g mol^{-1} molar mass of active materials, 0.5 Ω cm² area resistance and 0.79 V cell voltage. Similarly, to achieve the same cost target for non-aqueous systems, a combination of 100 g mol^{-1} molar mass of active materials, $2.5 \ \Omega \ cm^2$ area resistance, 3.0 V cell voltage, 0.2 salt ratio and 3.3 mol kg^{-1} specific amount of active material is required [16].

However, these conditions were not based on real systems and are still well beyond the foreseeable capabilities of most non-aqueous flow batteries. Techno-economic analyses shall consider real systems other than conventional chemistries that are emerging in both aqueous and non-aqueous electrolytes, including those organic-based and hybrid systems using different architectures. This analysis was attempted to evaluate the feasibility of these emerging systems to meet the cost target and to predict their technological prospects for energy storage applications. It is important to define further improvements required for larger cost reduction with the recent advances and mass productions of functional materials. This analysis is underpinned by a fact-based cost model, in which performance and cost parameters based on real systems are used. The capital costs of these resulting flow batteries are compared and discussed, providing suggestions for further improvements to meet the ambitious cost target for more effective market penetration in longterm.

2. Methodologies

2.1. The relevant RFB systems and grid storage applications

The proposed cost models are similar to previous models developed by Brushette and co-workers [16], taking account of raw active materials, solvents, cell components and performances based on well-known chemistries of different architectures (Fig. 1). This includes three commercial aqueous systems: acidic all-vanadium (all-V), acidic zinc-bromine (Zn-Br) and alkaline zinc ferricyanide (Zn-Fe(CN)₆); and six new organic based systems using aqueous (neutral viologen-TEMPO (Vi-TEMPO), acidic anthraquinone-bromine (AQ-Br) and alkaline phenazine-ferricyanide (Ph-Fe(CN)₆) and non-aqueous electrolytes (fluorenone-DBMMB (FL-DBMMB), all-metallocene (all-metallocene) and lithium-TEMPO (Li-TEMPO) [26–34]. The selections of emerging chemistries tended to be based on the citations of their first research documents in the relevant categories.

In addition, the model considers the actual cell settings, performances (e.g. cell voltages (*V*), efficiencies (η)) and architectures (true and hybrid) that were documented in the referenced articles (summarized in Table 1). For further cost reductions of these systems, the performances of the existing flow batteries need to be further improved in terms of usable active species concentrations, discharge voltages, number of electron-transfers and active material costs. The costs of organic active materials and non-aqueous solvents were assumed to be USD\$ 5 kg⁻¹ and USD\$ 3 L⁻¹, respectively, taking account of large volume productions. The costs of salts were USD\$ 0.1 kg⁻¹ and USD\$ 5 kg⁻¹ for aqueous and non-aqueous electrolytes [16,35]. In this model, electrolyte formulations were also simplified to only containing active

Table 1

The specifications and performances of the nine selected aqueous and non-aqueous now batteries evaluated in this techno-economic analysis [2
--

-		-		•	
Flow batteries	Chemicals	Relative molecular mass, Mr (g mol ⁻¹)	Current density, j (mA cm ⁻²) Columbic Efficiency, <i>CE</i> (%) Concentration, c (mol L ⁻¹) Discharge voltage, U (V) Number of electrons, N	Cost, \overline{C} (USD kg ⁻¹)	References
all-V	Vanadium	50.94	<i>j</i> : 60 / <i>CE</i> : 86 <i>c</i> : 1.5	23.6	J. Mater. Chem. A, 2015, 3,
			U : 1.4 / N : 1		16913
Zn-Br	Zinc (-)	65.38	j: 30 / CE: 95	2.3	Handbook of Batteries,
	Bromine (+)	159.82	c (-): 3 / c (+): 3	2.3	Chapter 37, McGraw-
			U : 1.6 / N : 2		Hill, 2002, 1-20.
Zn-Fe(CN) ₆	Zinc (-)	65.38	j: 20 / CE: 99	2.3	Joule, 2019, 3,
	Ferricyanide (+)	212	c (-): 0.12 / c (+): 0.8 U: 1.6 / N: 2	2.3	2255–2267
Vi-TEMPO	BTMAP-viologen (-)	360.22	j: 40 / CE: 99	5	Chem., 2019, 5,
	4-[3-(trimethylammonio)propoxy]-2,2,6,6- tetramethylpiperidine-1-oxyl (TMAP-TEMPO) chloride (+)	272.25	c (-): 0.1 / c (+): 0.1 U: 1.0 / N: 1	5	1861–1870
AO-Br	9.10-anthraquinone-2.7-disulphonic acid (-)	368.3	i: 500 / CE: 99	5	Nature, 2014, 505,
	Hydrobromic acid (+)	80.9	c (-): 1 / c (+): 3 U: 0.6 / N: 2	0.5	195–198
Ph-Fe(CN) ₆	7,8-dihydroxyphenazine-2-sulfonic acid (-)	292.3	j: 100 / CE: 99	5	Nat. Energy, 2018, 3,
	Ferricyanide (+)	348.8	c (-): 1.4 / c (+): 0.6 U : 1.3 / N : 2	2.3	508-514
FL-DBMMB	2,1,3-benzothiadiazole (-)	136.17	j: 40 / CE: 90	5	ACS Energy Lett. 2017,
	2,5-di-tert-butyl-1-methoxy-4-[2'- methoxyethoxy]benzene (+)	222.11	<i>c</i> (-): 0.1 / <i>c</i> (+): 0.1 <i>U</i> : 2 / <i>N</i> : 1	5	2, 5, 1156–1161
all-metallocene	Cobaltocene (-)	189.1	j: 1.5 / CE: 95	5	Energy Environ. Sci.,
	Ferrocene (+)	186	<i>c</i> (-): 0.1 / <i>c</i> (+): 0.1 <i>U</i> : 1.7 / <i>N</i> : 1	5	2017,10, 491-497
Li-TEMPO	Lithium (-)	6.9	<i>j</i> : 5 / <i>CE</i> : 99	7	Adv. Mater. 2014, 26,
	2,2,6,6-tetramethylpiperidine-1-oxyl (TEMPO) (+)	156.3	c (-): 2 / c (+): 0.1 U: 3 / N: 1	5	7649–7653



Fig. 2. The capital costs ((a) – (c)) and volumes ((d) – (f)) of aqueous and non-aqueous flow battery systems used in different applications (Application 1: solar energy integration ($2 \text{ MW} \times 6 \text{ h}$), Application 2: industrial load shifting ($5 \text{ MW} \times 4 \text{ h}$), Application 3: rural microgrid-households ($5 \text{ kW} \times 8 \text{ h}$), Application4: demand charge management ($10 \text{ MW} \times 11 \text{ h}$). Different colours in the bar chart represent different application scenarios.

materials, salts and solvents without specific additives, except the costly sequestration agents for bromine (USD $$28 \text{ kg}^{-1}$) [36] systems.

The power ratings and discharge durations vary with applications. For redox flow batteries, typical applications are solar energy integration (e.g. 2 MW \times 6 h), industrial load shifting (e.g. 5 MW \times 4 h), rural microgrid-households (e.g. 5 kW \times 8 h) and demand charge management (e.g. 10 MW \times 11 h), as defined by Görtz *et al.*[37,38]. Although storage capacities are often proportional to the electrolytes (in true flow batteries), the cell voltages and power densities are associated with the chemistries and cell settings (sizes, architecture and electrolytes). For all aforementioned systems, it is assumed that commercial Regenesys® type flow cell stacks (Fig. S1) with adjusted architectures (e.g. hybrid) were used in this cost model [37], since there was no standardised system available for direct comparison in the existing literatures. The component costs are based on previous works and current market prices as summarized in Table S1.

Key assumptions are as follows:

- (1) The cost of organic active materials and non-aqueous solvents are the same for all systems.
- (2) The types of component materials (including electrode and membrane) are identical for all systems (but in different architectures/configurations).
- (3) Commercial Regenesys® type flow cell stacks with adjusted architectures (e.g. hybrid) are used for all systems.
- (4) Electrolyte formulations are simplified to active materials, salts and solvents without additives (except bromine sequestration agents).
- (5) Crossover of active species across membranes and electrolyte leakage are not allowed.
- (6) Battery performances are independent from mass transport phenomena, current distribution (including shunt current), temperature and viscosity.
- (7) Electrical power is proportional to the number of stack, regardless of electrical/electrolyte connections.
- (8) Number of stacks can be a fraction instead of whole number.

(9) Capital cost of flow battery system does not include any cost associated with construction, labour, depreciation, overhead, profit and warranty.

2.2. The components and capital cost evaluation methods for RFB systems

The capital cost of flow battery includes the cost components of cell stacks (electrodes, membranes, gaskets and bolts), electrolytes (active materials, salts, solvents, bromine sequestration agents), balance of plant (BOP) (tanks, pumps, heat exchangers, condensers and rebalance cells) and power conversion system (PCS). Electrode materials includes bipolar plates, end-plates and graphite felts. The total costs of flow battery ($C_{\rm RFB}$) are expressed in terms of \$ (kW h)⁻¹ through dividing the costs of all these components ($C_{\rm stack}$, $C_{\rm electrolytes}$, $C_{\rm BOP}$ and $C_{\rm PCS}$) by the required energies of the applications ($E_{\rm total}=P \times t_{\rm discharge}$, where $P=V_{\rm discharge} \times t_{\rm discharge}$). It is assumed that power losses have been taken account as form of discharge voltages obtained from the source data.

$$C_{RFB} = C_{stack} + C_{electrolytes} + C_{BOP} + C_{PCS}$$

$$(2.1)$$

$$C_{stack} = C_{electrodes} + C_{membranes} + C_{gaskets} + C_{bolts}$$
(2.2)

$$C_{electrolytes} = C_{actives} + C_{salts} + C_{solvents} + C_{BSA}$$
(2.3)

$$C_{electrolytes} = m_{actives}\bar{C}_{actives} + m_{salts}\bar{C}_{salts} + V_{solvents}\bar{C}_{solvents} + m_{BSAs}\bar{C}_{BSA}$$
(2.4)

$$C_{BOP} = C_{tanks} + C_{pumps} + C_{heatexch} + C_{condenser} + C_{rebalancecells}$$
(2.5)

$$C_{BOP} = A_{tanks}\bar{C}_{tanks} + n(2\bar{C}_{pump} + \bar{C}_{heatexch} + \bar{C}_{condenser}) + E_{total}\bar{C}_{rebalancecells}$$
(2.6)

$$C_{PCS} = C_{BMS} = n\bar{C}_{BMS} \tag{2.7}$$

where the electrolyte cost (C_{RFB}) consists of the masses of active materials (m_{actives}), salts (m_{salts}) and sequestration agents (m_{BEP}) multiplying their cost per kg (\bar{C}) and the volume of solvents (V_{solvents} , unit: m³ multiplying their cost per volume ($\bar{C}_{\text{solvents}}$). The masses of active



Fig. 3. The cost breakdowns of aqueous and non-aqueous flow battery systems used in Application 1 (2 MW \times 6 h).

materials were calculated by Faradays law, taking account of coulombic efficiencies in the charge-discharge processes. The cost of tank (C_{tanks}) is estimated by multiplying the exposed areas to the electrolytes ($A_{\text{tanks}}=2\pi(1+V_{\text{solvents}}/1000\pi)$, unit: m²) with the cost of building (insulating) material per m² (\bar{C}_{tanks}). The cost of rebalance cell depends on the total power output E_{total} , and obtained by multiplying with its cost per kW ($\bar{C}_{\text{rebalance cells}}$). The costs of the other BOP items and battery management system both are proportional to the number of cell stacks, however two pumps are required in each stack. \bar{C}_{pump} , $\bar{C}_{\text{heat exch}}$, $\bar{C}_{\text{condenser}}$ and \bar{C}_{BMS} are the cost of each pump, heat exchanger, condenser and battery management system (BMS), respectively.

2.3. The calculation of relative sensitivity index of components

The relative sensitivity index of components was defined by Eq. (2.8) [39]:

Relatives
ensitivity index =
$$abs(\frac{df}{dx/BCP} \times \frac{x_0}{f_0})$$
 (2.8)

where *BCP* is the base case point, *x* is the cost component variable, *f* is the capital cost function, x_0 is the variable at the base case, and f_0 is the overall capital cost at the base case. The relative sensitivity index is the measure of the rate of change of the capital costs for a particular variable at the base case. It is expected that more influences of these variables on the capital costs with higher relative sensitivity indexes.

3. Results and discussion

3.1. Conventional and emerging flow batteries for grid storage applications

To-date, redox flow batteries are mainly used for different grid-scale

applications, which have different power ratings and discharge durations [4]; and are assumed as follows: solar energy integration (as Application 1: e.g. 2 MW \times 6 h), industrial load shifting (as Application 2: e.g. 5 MW \times 4 h), rural microgrid-households (as Application 3: e.g. 5 $kW \times 8$ h) and demand charge management (as Application 4: e.g. 10 MW \times 11 h) [37,38]. In this techno-economic analysis, the capital costs and volumes of cell stacks and electrolytes were evaluated for existing commercial and emerging systems using aqueous and non-aqueous electrolytes as shown in Fig. 2. It should be noted that the y-axes are not adjusted to the same scale. Regardless of systems, the capital costs per kW h are observed to be lower for applications with higher E/P ratios (i.e. smaller power rating and longer duration). This is because small power implies fewer cells or stacks required but energy (content or capacity) increases with the amounts of the electrolytes. By increasing the discharge durations, lower capital costs per kW h were observed as the increased electrolyte cost fractions offset the expenses per kW h of the cell stacks [18].

Existing commercial systems are all based on aqueous electrolytes, three of the commonest systems are evaluated in this cost model: (1) allvanadium (all-V), (2) zinc-bromine (Zn-Br) and (3) zinc ferricyanide (Zn-Fe(CN)₆) (Fig. 2a and d). The capital costs of all-V and Zn-Br systems were estimated to be USD\$ $170 - 580 (kW h)^{-1}$ and comparable with previous reports (USD\$ 350 — 600 (kW h)⁻¹ at E/P = 4) [40,41], which are still higher than the DoE cost target (USD\$ < 100 (kW h)⁻¹). Despite the lower cost of active materials (Zn: USD 2.3 kg⁻¹), the costs of these zinc-based hybrid systems were higher than that of all-V system. This was because more cell stacks are required for the same power output due to the uses of lower current densities ($< 50 \text{ mA cm}^{-2}$) associated with the electrodeposition processes. However, bromine electrolytes involve sequestration agents (USD\$ 28 kg⁻¹) that are as expensive as vanadium (USD\$ 24 kg⁻¹). For the case of Zn-Fe(CN)₆ batteries, although the electrolyte costs were as low as USD\$ 13.75 (kW h)⁻¹ (E/P = 4), the current densities were even smaller (20 mA cm⁻²) in alkaline



Fig. 4. The cost projections of aqueous and non-aqueous flow battery systems under the influences of (a, b) current density and (c, d) duration used in Application 1 (2 MW \times 6 h).

electrolytes to avoid dendritic/mossy electrodeposits, resulting in high stack costs of more than USD\$ 200 (kW h) $^{-1}$.

For alternative systems, three emerging aqueous batteries were evaluated (Fig. 2b and e): (1) all-organic (Vi-TEMPO), (2) organicinorganic (halogen) (AQ-Br) and (3) organic-inorganic (metal) (Ph-Fe (CN)₆). Vi-TEMPO battery was the only system more costly than the existing commercial systems. Similar to many all-organic aqueous batteries, this battery tends to have lower cell voltages (< 1.4 V) and current densities ($< 80 \text{ mA cm}^{-2}$), leading to one of the highest stack costs in aqueous systems [28–30]. Although organic active materials are reasonably low-cost in long-term (USD 5 kg⁻¹), the electrolyte costs were still higher than their vanadium counterparts. This was mainly attributed to the relatively large molecular weight of these organic molecules (several times larger than metals) [11,42]. Unlike other organic material (e.g. quinone, ketone) [43], the reactions of the all-organic Vi-TEMPO battery only involves one electron-transfer and yields a lower cell voltage of c.a. 1 V [29]. On the other hand, the selected organic-inorganic batteries appear to have the lowest capital costs (USD\$115 - 339 (kW h)⁻¹), this can be explained by their significant cost reduction in stack number due to their impressive current densities ($\geq 100 \text{ mA cm}^{-2}$). It is worth noting that the costs per kW h of these electrolytes (organic-inorganic systems) were comparable with those using low-cost metals (e.g. Zn and Fe) when no expensive additive or complexing agent (e.g. bromine sequestration agent) is used. Despite the negligible cost of water solvent (USD\$ 0.002 L^{-1}) [37], the electrolyte volumes are mainly proportional to the usable concentrations but also their energy contents in terms of cell voltages and electron-transfers regardless of applications, much larger than the unit energy volume of Li-ion batteries (5 L (kW h)⁻¹) (Fig. 2d to f) [44]. Due to the low concentration (0.1M) and high molecular weight (272 and 360 g mol^{-1}), all-organic Vi-TEMPO battery has the largest volume (per kW h) as shown in Fig. 2e.

In addition to aqueous flow batteries, three representative systems using non-aqueous electrolytes were considered: FL-DBMMB, all-metallocene and lithium-TEMPO batteries (Fig. 2c and f) [32–34]. These non-aqueous batteries exhibited higher cell voltages (≥ 1.7 V) than aforementioned aqueous systems [45], the capital costs of these system remained prohibitively expensive of over USD\$ 800 (kW h)⁻¹. The main reasons were the low usable concentrations (0.1 M) of active species, hence large volume of higher cost non-aqueous electrolytes (USD\$ 3 L⁻¹) were used instead of water (Fig. 2f). The costs of these non-aqueous electrolytes were up to USD\$ 9400 (kW h)⁻¹ but less volume can be achieved with active materials of larger energy contents (i.e. cell voltage, electron-transfers) and smaller molecular weights as the given case of Li-TEMPO electrolytes (USD\$ 1237 (kW h)⁻¹ at E/P = 4 (Fig. 2c and f). For all-metallocene and Li-TEMPO batteries, stack costs were the other main contributors of high capital costs, since more stacks were required due to the use of relatively low current densities (\leq 5 mA cm⁻²). However, FL-DBMMB flow battery demonstrated the use of 40 mA cm⁻², resulting in comparable stack costs with some aqueous systems of similar current densities.

3.2. Cost breakdowns of redox flow batteries

The capital costs of flow batteries have been simplified into several cost components of cell stacks (electrodes, membranes, gaskets and bolts), electrolytes (active materials, salts, solvents, bromine sequestration agents), balance of plant (BOP) (tanks, pumps, heat exchangers, condensers and rebalance cells) and power conversion system (PCS). Using solar energy integration (e.g. $2 \text{ MW} \times 6 \text{ h}$) as an example, the cost breakdowns of conventional and emerging flow batteries are shown in Fig. 3. It can be seen that the total capital costs are lower using aqueous systems (USD\$ < 700 (kW h)⁻¹), especially for those organic-inorganic systems that have relatively low stack costs due to the uses of relatively large current densities ($\geq 100 \text{ mA cm}^{-2}$) [30,31]. For most aqueous flow batteries with smaller current densities, the cost proportions of membrane are significant, up to more than 40 %. However, the case of AQ-Br system is exceptional as they effectively reduce the use of cell stacks due to their impressive current densities (500 mA cm⁻²), membrane only accounted for about 10 % of the total capital cost. For most aqueous systems, the costs of active materials and sequestration agents are the other major cost components, especially when bromine was used as active species. Compared to membranes and electrolyte constituents, the cost proportions of the other components, such as electrodes and



Fig. 5. The cost projections of aqueous and non-aqueous flow battery systems with lower-cost membranes (a, b) and electrode materials (c, d) used in Application 1 (2 MW \times 6 h).

ancillary components, were even less significant.

For non-aqueous emerging systems, the capital costs are prohibitively high for both cell stacks (USD\$ > 150 (kW h)⁻¹) and electrolytes $(USD\$ > 400 (kW h)^{-1})$. Since non-aqueous electrolytes are less conductive than water [17], it is challenging to develop systems with comparable current densities ($\geq 100 \text{ mA cm}^{-2}$) to their aqueous counterparts, leading to higher stack costs. Unless obtaining relatively high current densities as in the case of FL-DBMMB battery, the cost fractions of stack component materials, particularly membranes and electrodes, are the most significant as in aqueous systems. Otherwise, non-aqueous solvents are likely the highest cost components of the flow batteries depending on the usable concentrations of their active species, given the fact that the three selected non-aqueous systems only use active species of 0.1 M concentrations. Despite the significant costs of non-aqueous solvents, the active material costs of these systems were relatively low $(USD\$ < 50 (kW h)^{-1})$. The cost fractions of other components (pump, heat exchange and battery management system) were not significant but are entirely dependent on the required stack number.

3.3. Further cost reductions for existing flow batteries

Previous sections evaluated the capital costs of existing flow batteries for grid-scale applications. Further cost reduction may be achieved with these systems if improved operating conditions and lower-cost components are possible in the future. Regardless of systems, increasing current density is known to be an effective approach to reduce the stack number for the same power ratings (Fig. 4a and b) [39]. The use of higher current density is subjected to reaction kinetics, mass transport of active species, resistance of systems and quality of electrodeposits [46]. As demonstrated in recent aqueous and non-aqueous systems, it is feasible to increase the current densities of up to 500 mA cm⁻² (e.g. AQ-Br [30]) and 40 mA cm⁻² (e.g. FL-DBMMB [32]), respectively, for true flow batteries, although hybrid batteries tend to use lower current densities due to the electrodeposition reactions [12].

This approach is particularly effective for systems of using low-cost electrolytes (solvents and constituents), e.g. Zn-Fe(CN)₆ (hybrid) and Ph-Fe(CN)₆ batteries. However, the effect was less pronounced when current densities increased to more than 100 mA cm⁻² (Fig. 4a) and 20 mA cm⁻² (Fig. 4b) for aqueous and non-aqueous systems, respectively. Certain flow batteries may meet the DoE cost target (USD\$ 100 (kW h)⁻¹) within reasonable ranges of current densities (e.g. Ph-Fe(CN)₆ at *c. a.* 240 mA cm⁻²). For their non-aqueous counterparts, their capital costs remained prohibitively high (USD\$ > 480 (kW h)⁻¹), indicating that increasing current densities was still not sufficient for commercial reality.

In addition to current densities, it is also known that the capital costs of flow batteries are lower at higher E/P ratios, characterized by smaller power ratings and longer discharge durations. However, these are very dependent on the applications or/and the demands of the end-users, thus very limited rooms for further adjustments. Similar to increasing current densities, cost reductions can also be achieved by increasing the discharge durations (Fig. 4c and d), especially for those using low-cost electrolytes or expensive stacks. As a form of energy storage capacities, discharge durations increase only with the amount of electrolytes, the increased electrolyte cost fractions tend to offset the expenses of the cell stacks, leading to lower capital costs per kW h [25]. When the discharge durations were increased up to 14 h, the capital cost of aqueous Ph-Fe(CN)₆ battery would be lower than USD\$ 110 (kW h)⁻¹ (Fig. 4c). Thus, increasing current densities and discharge durations (or E/P ratios) are both effective approaches for further cost reductions.

Further capital cost reductions can be achieved with lower-cost membranes (Fig. 5a and b) and electrode materials (Fig. 5c and d), the current prices are up to USD\$ 500 m⁻² and USD\$ 77 m⁻², respectively [37]. Ha and Gallagher [21] suggested that the future cost of membranes and electrodes may be reduced to USD\$ 50 m⁻² and USD\$ 20 m⁻² with high-volume productions of 20,000 stack per year, suggesting drastic drops in membrane cost of up to 90 % are expected in the future. As one of the emerging membranes, the cost of polybenzimidazole (PBI)



Fig. 6. The cost projections of aqueous and non-aqueous flow battery systems under the influences of usable active material concentrations (a, b), number of electron-transfers (c, d) and discharge cell voltages (e, f) used in Application 1 (2 MW \times 6 h).

membrane has been reported to be as low as USD\$ 40 m^{-2} but not yet commercialized at large-scale [47]. Furthermore, future membranes shall also be both conductive and selective without sacrificing the cell performances.

Taking account that cost fractions of membranes are among the largest compared with other cell components (in some systems up to 40 %, such as all-metallocene, Li-TEMPO, Zn-Fe(CN)₆, Vi-TEMPO and Zn-Br), cost reductions are more pronounced with future state membranes than with electrodes. Thus, it may also be useful in exploring new hybrid flow battery systems that may eliminate membrane/ separator. For instance, the capital cost of a membraneless RFB (Zn-pBQ) can be lower than USD\$ 100 (kW h)⁻¹ (Application 4: 10 MW × 11 h) in this cost model (Table S8).

With the use of low-cost membranes and electrodes (at future state costs), the capital cost of aqueous Ph-Fe(CN)₆ battery was estimated to be USD\$ 107 (kW h)⁻¹ and USD\$ 154 (kW h)⁻¹, respectively (Fig. 5a and c). When these materials are used simultaneously, the capital cost is further reduced to USD\$ 92 (kW h)⁻¹, reaching the cost target (USD\$ 100 (kW h)⁻¹) without further improving the cell performances. For non-aqueous systems, more number of stacks (higher stack cost) is often

expected due to the uses of lower current densities (< 20 mA cm⁻²). It is known that membranes and electrodes are major stack components, notable cost reductions were still observed with these materials at future state costs, except for the case of FL-DBMMB battery (Fig. 5b and d). However, the solvent costs of the three non-aqueous systems were still too high (USD\$ > 400 (kW h)⁻¹), suggesting that the cost reductions of electrode and membrane materials are not sufficient for competitive systems.

3.4. Pathways to lower-cost flow batteries based on existing systems

Despite the possibilities of further cost reductions for existing flow batteries, explorations of more competitive systems need to be continued and facilitated with new chemistries of enhanced properties in both aqueous and non-aqueous electrolytes. Based on the existing architectures, capital costs of the aforementioned flow batteries were evaluated with several variables, i.e. usable active material concentrations (Fig. 6a and b), number of electron-transfers (Fig.s 6c and d) and discharge cell voltages (Fig. 6e and f). These variables are only associated with active materials and are independent from engineering Table 2

Sensitivities of capital cost of aqueous and non-aqueous flow batteries at different design parameters and material costs (Application 1: $2 \text{ MW} \times 6 \text{ h}$)

Sensitivities RFBs	Current density	Duration	Membrane cost	Electrode cost	Usable species concentration	Discharge voltage	Number of electron-transfers
all-V	0.7298	0.8132	0.6327	0.5323	0.5014	0.9425	0.5914
Zn-Br	0.7131	0.7820	0.6347	0.5158	0.5003	0.9659	0.6105
Zn-Fe(CN) ₆	0.9173	0.9653	0.7522	0.5255	0.5012	0.9641	0.5084
Vi-TEMPO	0.7371	0.8034	0.6363	0.5331	0.5123	0.9712	0.5977
AQ-Br	0.5403	0.5785	0.5252	0.507	0.5006	0.9525	0.6822
Ph-Fe(CN) ₆	0.7187	0.8182	0.6271	0.5312	0.5048	0.9163	0.5464
FL-DBMMB	0.5284	0.5454	0.5186	0.5052	0.6289	0.9902	0.8949
all-metallocene	0.8157	0.8612	0.6736	0.5403	0.5277	0.9977	0.5630
Li-TEMPO	0.7403	0.7973	0.6572	0.5179	0.5922	0.9877	0.6018

approaches. Further improvements are challenging unless organic-based active materials are considered. Also, enhanced properties are required for both negative and positive electrode reactions simultaneously. However, cost reductions of improving these design variables are only effective in certain flow batteries. For instance, increasing usable active material concentrations and number of electron-transfers reduce the electrolyte volumes, cost reductions are insignificant for aqueous batteries considering the negligible cost of water (Fig. 6a and c). By contrast, cost reductions are more pronounced with non-aqueous systems with larger cost fractions of electrolytes (or with higher current densities) (Fig. 6b and d). The estimated cost of FL-DBMMB battery was down to USD\$ 614 (kW h)⁻¹(up to 40 % reduction) and USD\$ 573 (kW h)⁻¹(up to 37 % reduction) if 3 M usable concentrations and 6 electron-transfers were realized, respectively.

Increasing discharge voltage is another effective approach in reducing the capital costs of flow batteries (Fig. 6e and f). This is because increasing discharge voltage not only implies fewer stacks but also reduces the amount of the electrolytes. However, cell voltages are subjected to the selections of redox couples and the stability windows of solvents. Despite the stability window of water is only 1.23 V [48], some aqueous systems have been demonstrated to yield the cell voltages of more than 2 V by suppressing the gas evolutions [49,50]. If the cell voltage of Ph-Fe(CN)₆ battery (1.3 V) was increased to 2 V, the capital cost could drop to the DoE target (from USD\$ 168 $(kW h)^{-1}$) with cost reductions of greater than 40 % for both stacks and electrolytes (Fig. 6e). On the other hand, most non-aqueous solvents have stability windows of more than 4 V but suitable redox pairs with high cell voltages (i.e. > 2 V) remain rare (except hybrid systems with lithium anodes). Nevertheless, it is still realistic to obtain cell voltages of 3 and 4 V for true and hybrid flow batteries, respectively. Based on these targets, the capital costs of the three non-aqueous batteries are still significantly higher than USD\$ 400 (kW h)⁻¹, requiring simultaneous measures to meet the DoE cost target.

4. Conclusions

In this techno-economic analysis, the capital costs of existing commercial and emerging systems were evaluated using aqueous and nonaqueous electrolytes. Regardless of systems, the capital costs per kW h are lower for applications with higher E/P ratios (i.e. smaller power rating and longer duration). Existing commercial flow batteries (all-V, Zn-Br and Zn-Fe(CN)₆ batteries; USD $\$ > 170 \text{ (kW h)}^{-1}$)) are still far beyond the DoE target (USD\$ 100 (kW h)⁻¹), requiring alternative systems and further improvements for effective market penetration. Compared to conventional redox couples, organic active materials offer possibilities in terms of improved properties (e.g. solubilities and multielectron transfers) and are expected to be low-cost in long term (USD\$ 5 kg⁻¹). However, organic-based batteries (e.g. Vi-TEMPO battery) do not necessarily guarantee lower capital costs because of their relatively large molecular weights (> 100 g mol^{-1}) and lower cell voltages (aqueous: <1.3 V; non-aqueous: < 2.2 V), also some do not involve multi-electron transfers. If no expensive additives or complexing agents are used, some aqueous organic-inorganic batteries (e.g. Ph-Fe(CN)₆ battery)

have the lowest capital cost (USD\$ ${\sim}115~(kW~h)^{-1}$) owing to their impressive current densities (${\geq}~100~mA~cm^{-2}$). By contrast, non-aqueous batteries are all prohibitively expensive (USD\$ ${>}800~(kW~h)^{-1}$) as these systems use relatively low concentrations (${<}~0.5~M$) and current densities (${<}~20~mA~cm^{-2}$), implying more amounts of electrolytes and stacks.

Based on the existing systems, further cost reductions were examined with improved operating conditions and lower-cost stack components. The sensitivities of capital costs to different variables are summarized in Table 2. Discharge voltage, duration and current density were observed to have the highest influences on cost reductions (higher relative sensitivity indexes), hence further improvements are required preferably. Regarding the operating conditions, increasing current densities and discharge durations (or E/P ratios) are effective approaches for overall cost reductions. Although current densities can be improved through engineering approaches (e.g. minimizing resistances), the latter are mainly dependent on the applications. As an alternative, the stack costs can be reduced effectively with the expected drops of membrane and electrode material costs (over 70 %) benefiting from high-volume productions. Since membranes account for the largest cost fractions of stack components and have higher relative sensitivity indexes, cost reductions are more pronounced than those with electrodes. With these materials at future state costs, the capital cost of aqueous Ph-Fe(CN)₆ battery (USD\$ 93 (kW h)⁻¹) is lower than the DoE cost target without further improving the cell performances. The aforementioned approaches are also effective for non-aqueous batteries, their capital costs $(USD\$ > 400 (kW h)^{-1})$ remain too high, especially for those using low active material concentrations with large volumes of non-aqueous solvents.

To facilitate the exploration of more competitive systems, capital cost evaluations of new chemistries with enhanced properties (usable active material concentrations, number of electron-transfers and cell voltages) were evaluated using existing batteries/ architectures, which are only associated with active materials and independent from engineering approaches. Increasing usable active material concentrations and number of electron-transfers effectively reduce the electrolyte volumes, and hence their costs. Despite significant savings for non-aqueous systems (down to USD\$ < 410 (kW h)⁻¹ at 6 e⁻ transfers), this approach was not pronounced for their aqueous counterparts due to the negligible cost of water. Unlike the other variables, increasing discharge voltage is among the most effective approach in cost reduction, as it not only implies fewer stack but also reduce the amount of the electrolytes. It is also found that the cost reductions are briefly proportional to their capital cost reduction. Even though impressive properties have been obtained, it may not be sufficient to meet the DoE cost targets unless different measures are taken simultaneously. Based on the sensitivity data, quantitative iterations were conducted by considering the aforementioned parameters. With the modest improvements to these variables, realistic design pathways for aqueous and non-aqueous flow batteries (true and hybrid) towards the cost targets are suggested by avoiding ineffective cost-cutting strategies (Table 2). It can be seen that competitive systems are still realistic from the current status of aqueous flow batteries, while their non-aqueous counterparts remain challenging unless tremendous improvements (e.g. higher current density, wider voltage window) have been made on several aspects. The dedicated focus on capital cost estimations of existing systems may provide important insight for designing economically viable flow batteries.

CRediT authorship contribution statement

L. Tang: Data curation, Writing – original draft. P. Leung: Writing – review & editing, Conceptualization, Methodology, Software. M.R. Mohamed: Visualization, Investigation. Q. Xu: Conceptualization, Methodology, Software. S. Dai: Visualization, Investigation. X. Zhu: Supervision. C. Flox: Conceptualization, Methodology, Software. A.A. Shah: Supervision. Q. Liao: Supervision.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

This work is supported by the Innovative research group project of National Natural Science Foundation of China (No. 52021004), the Funds for Chongqing Talents Plan (No. CQYC2021059563) and the Fundamental Research Funds for the Central Universities (No. 2021CDJQY-027). C. Flox acknowledges financial support by Ministery of Science Spain, AEI Severo Ochoa Grant CEX2019-000917-S and European Commission under the grant MSCA-IF-EF-ST, proposal number 101026162.

Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.electacta.2022.141460.

References

- T.M. Gür, Review of electrical energy storage technologies, materials and systems: challenges and prospects for large-scale grid storage, Energy Environ. Sci. 11 (2018) 2696–2767, https://doi.org/10.1039/c8ee01419a.
- [2] S. Hameer, J.L. van Niekerk, A review of large-scale electrical energy storage, Int. J. Energy Res. 39 (2015) 1179–1195, https://doi.org/10.1002/er.3294.
- [3] DOE Global Energy Storage Database, Sandia National Laboratories and DOE Office of Electricity, US Department Energy (2019). https://www.osti.gov/servlets/pur 1/1642605.
- [4] M.A. Rahman, J.H. Kim, S. Hossain, Recent advances of energy storage technologies for grid: A comprehensive review, Energy Storage (2022), https://doi. org/10.1002/est2.322.
- [5] M. T. P. Ralon, A. Ilas, H. Diaz-Bone, K.-P. Kairies, Electricity storage and renewables: costs and markets to 2030 *Int. Renew. Energy Agency* Abu Dhabi (2017) https://www.irena.org/publications/2017/oct/electricity-storage-and-rene wables-costs-and-markets.
- [6] Martin Brudermüller, B.S. Dominic Waughray, A vision for a sustainable battery value chain in 2030: unlocking the full potential to power sustainable development and climate change mitigation, World Economic Forum (2019). https://www3.we forum.org/docs/WEF_A_Vision_for_a_Sustainable_Battery_Value_Chain_in_2030_Re port.pdf.
- [7] J.S. Edge, et al., Lithium ion battery degradation: what you need to know, Phys. Chem. Chem. Phys. 23 (2021) 8200–8221, https://doi.org/10.1039/d1cp00359c
- [8] V. Etacheri, R. Marom, R. Elazari, G. Salitra, D. Aurbach, Challenges in the development of advanced Li-ion batteries: a review, Energy Environ. Sci. 4 (2011), https://doi.org/10.1039/c1ee01598b.
- W. Wang, et al., Recent Progress in Redox Flow Battery Research and Development, Adv. Funct. Mater. 23 (2013) 970–986, https://doi.org/10.1002/ adfm.201200694.
- [10] G.L. Soloveichik, Flow Batteries: Current Status and Trends, Chem. Rev. 115 (2015) 11533–11558, https://doi.org/10.1021/cr500720t.
- [11] J. Winsberg, T. Hagemann, T. Janoschka, M.D. Hager, U.S. Schubert, Redox-flow batteries: from metals to organic redox-active materials, Angew Chem Int Ed Engl 56 (2017) 686–711, https://doi.org/10.1002/anie.201604925.
- [12] A. Khor, et al., Review of zinc-based hybrid flow batteries: From fundamentals to applications, Mater. Today Energy 8 (2018) 80–108, https://doi.org/10.1016/j. mtener.2017.12.012.

- [13] Kendall Mongird, V. V., Jan Alam, Charlie Vartanian, Vincent Sprenkle, Richard Baxter. Grid Energy Storage Technology Cost and Performance Assessment. U.S. Department of energy. (2020),https://www.pnnl.gov/sites/default/files/media/f ile/Final%20-%20ESGC%20Cost%20Performance%20Report%2012-11-2020.pdf.
- [14] M. Park, J. Ryu, W. Wang, J. Cho, Material design and engineering of nextgeneration flow-battery technologies, Nat. Rev. Mater. 2 (2016), https://doi.org/ 10.1038/natrevmats.2016.80.
- [15] P. Leung, et al., Recent developments in organic redox flow batteries: A critical review, J. Power Sources 360 (2017) 243–283, https://doi.org/10.1016/j. jpowsour.2017.05.057.
- [16] R. Dmello, J.D. Milshtein, F.R. Brushett, K.C. Smith, Cost-driven materials selection criteria for redox flow battery electrolytes, J. Power Sources 330 (2016) 261–272, https://doi.org/10.1016/j.jpowsour.2016.08.129.
- [17] L. Tang, et al., Future perspective on redox flow batteries: aqueous versus nonaqueous electrolytes, Curr. Opin. Chem. Eng. 37 (2022), https://doi.org/ 10.1016/j.coche.2022.100833.
- [18] R.M. Darling, K.G. Gallagher, J.A. Kowalski, S. Ha, F.R. Brushett, Pathways to lowcost electrochemical energy storage: a comparison of aqueous and nonaqueous flow batteries, Energy Environ. Sci. 7 (2014) 3459–3477, https://doi.org/10.1039/ c4ee02158d.
- [19] K Mongird, V. F., V Viswanathan, V Koritarov, P Balducci, B Hadjerioua, J Alam. Energy Storage Technology and Cost Characterization Report. (2019), https://en ergystorage.pnnl.gov/pdf/PNNL-28866.pdf.
- [20] M. Dieterle, et al., Life cycle assessment (LCA) for flow batteries: a review of methodological decisions, Sustain. Energy Technol. Assessments 53 (2022), https://doi.org/10.1016/j.seta.2022.102457.
- [21] S. Ha, K.G. Gallagher, Estimating the system price of redox flow batteries for grid storage, J. Power Sources 296 (2015) 122–132, https://doi.org/10.1016/j. jpowsour.2015.07.004.
- [22] T.D. Gregory, M.L. Perry, P. Albertus, Cost and price projections of synthetic active materials for redox flow batteries, J. Power Sources 499 (2021), https://doi.org/ 10.1016/j.jpowsour.2021.229965.
- [23] V. Viswanathan, et al., Cost and performance model for redox flow batteries, J. Power Sources 247 (2014) 1040–1051, https://doi.org/10.1016/j. jpowsour.2012.12.023.
- [24] Y.K. Zeng, T.S. Zhao, L. An, X.L. Zhou, L. Wei, A comparative study of all-vanadium and iron-chromium redox flow batteries for large-scale energy storage, J. Power Sources 300 (2015) 438–443, https://doi.org/10.1016/j.jpowsour.2015.09.100.
- [25] F.R. Brushett, M.J. Aziz, K.E. Rodby, On lifetime and cost of redox-active organics for aqueous flow batteries, ACS Energy Lett 5 (2020) 879–884, https://doi.org/ 10.1021/acsenergylett.0c00140.
- [26] K.J. Kim, et al., A technology review of electrodes and reaction mechanisms in vanadium redox flow batteries, J. Mater. Chem. A 3 (2015) 16913–16933, https:// doi.org/10.1039/c5ta02613j.
- [27] P. A. E. P.C. Butler, P.G. Grimes, S.E. Klassen, R.C. Miles, Handbook of, Batteries, b. D. L. a. T. B. R., Chapter 37-Zinc/Bromine Batteries, & third ed, 2002, pp. 1–20. M.-H. https://www.sandia.gov/ess-ssl/publications/SAND2000-0893.pdf.
- [28] Y. Chen, et al., A Stable and High-Capacity Redox Targeting-Based Electrolyte for Aqueous Flow Batteries, Joule 3 (2019) 2255–2267, https://doi.org/10.1016/j. joule.2019.06.007.
- [29] Y. Liu, et al., A Long-Lifetime All-Organic Aqueous Flow Battery Utilizing TMAP-TEMPO Radical, Chem 5 (2019) 1861–1870, https://doi.org/10.1016/j. chempr.2019.04.021.
- [30] B. Huskinson, et al., A metal-free organic-inorganic aqueous flow battery, Nature 505 (2014) 195–198, https://doi.org/10.1038/nature12909.
- [31] A. Hollas, et al., A biomimetic high-capacity phenazine-based anolyte for aqueous organic redox flow batteries, Nature Energy 3 (2018) 508–514, https://doi.org/ 10.1038/s41560-018-0167-3.
- [32] J. H. Wentao Duan, Jeffrey A. Kowalski, Ilya A. Shkrob, M. Vijayakumar, Eric Walter, Baofei Pan, Zheng Yang, Jarrod D. Milshtein, Bin Li, Chen Liao, Zhengcheng Zhang, Wei Wang, Jun Liu, Jeffrey S Moore, Fikile R. Brushett, Lu Zhang, Xiaoliang Wei, Wine-dark sea" in an organic flow battery: storing negative charge in 2,1,3-benzothiadiazole radicals leads to improved cyclability ACS Energy Lett. 2 (2017) 1156–1161, https://doi.org/10.1021/ acsenergylett.7b00261.
- [33] Y. Ding, Y. Zhao, Y. Li, J.B. Goodenough, G. Yu, A high-performance allmetallocene-based, non-aqueous redox flow battery, Energy Environ. Sci. 10 (2017) 491–497, https://doi.org/10.1039/c6ee02057g.
- [34] X. Wei, et al., TEMPO-based catholyte for high-energy density nonaqueous redox flow batteries, Adv Mater 26 (2014) 7649–7653, https://doi.org/10.1002/ adma.201403746.
- [35] G. Martin, L. Rentsch, M. Höck, M. Bertau, Lithium market research global supply, future demand and price development, Energy Storage Materials 6 (2017) 171–179, https://doi.org/10.1016/j.ensm.2016.11.004.
- [36] R.V. Adith, R.p. Naresh, K. Mariyappan, M. Ulaganathan, P. Ragupathy, An optimistic approach on flow rate and supporting electrolyte for enhancing the performance characteristics of Zn-Br₂ redox flow battery, Electrochim. Acta 388 (2021), https://doi.org/10.1016/j.electacta.2021.138451.
- [37] H.Y. Han. Cost assessment of organic redox flow battery for practical applications, University of Southampton Thesis, 2020.
- [38] Görtz, S. Battery energy storage for intermittent renewable electricity production: A review and demonstration of energy storage applications permitting higher penetration of renewables. (2015), http://www.diva-portal.org/smash/get/diva2: 818683/FULLTEXT01.pdf.

L. Tang et al.

- [39] M. Zhang, M. Moore, J.S. Watson, T.A. Zawodzinski, R.M. Counce, Capital Cost Sensitivity Analysis of an All-Vanadium Redox-Flow Battery, J. Electrochem. Soc. 159 (2012) A1183–A1188, https://doi.org/10.1149/2.041208jes.
- [40] M. Ulaganathan, et al., Recent advancements in all-vanadium redox flow batteries, Adv. Mater. Interfaces 3 (2016), https://doi.org/10.1002/admi.201500309.
- [41] K. Shin, J.-H. Lee, J. Heo, H.-T. Kim, Current status and challenges for practical flowless Zn–Br batteries, Curr. Opin. Electrochem 32 (2022), https://doi.org/ 10.1016/j.coelec.2021.100898.
- [42] X. Wei, et al., Materials and Systems for Organic Redox Flow Batteries: Status and Challenges, ACS Energy Lett. 2 (2017) 2187–2204, https://doi.org/10.1021/ acsenergylett.7b00650.
- [43] M. Pahlevaninezhad, et al., A nonaqueous organic redox flow battery using multielectron quinone molecules, J. Power Sources 500 (2021), https://doi.org/ 10.1016/j.jpowsour.2021.229942.
- [44] M. Ikezoe, et al., Development of High Capacity Lithium- Ion Battery for NISSAN LEAF, SAE Technical Paper Series (2012). https://www.sae.org/publications /technical-papers/content/2012-01-0664/.

- [45] K. Gong, Q. Fang, S. Gu, S.F.Y. Li, Y. Yan, Nonaqueous redox-flow batteries: organic solvents, supporting electrolytes, and redox pairs, Energy Environ. Sci. 8 (2015) 3515–3530, https://doi.org/10.1039/c5ee02341f.
- [46] X. Ke, J.I.D. Alexander, J.M. Prahl, R.F. Savinell, Flow distribution and maximum current density studies in redox flow batteries with a single passage of the serpentine flow channel, J. Power Sources 270 (2014) 646–657, https://doi.org/ 10.1016/j.jpowsour.2014.07.155.
- [47] Z. Li, Y.-C. Lu, Polysulfide-based redox flow batteries with long life and low levelized cost enabled by charge-reinforced ion-selective membranes, Nature Energy 6 (2021) 517–528, https://doi.org/10.1038/s41560-021-00804-x.
- [48] H. Zhang, X. Liu, H. Li, I. Hasa, S. Passerini, Challenges and strategies for highenergy aqueous electrolyte rechargeable batteries, Angew. Chem. Int. Ed. Engl. 60 (2021) 598–616, https://doi.org/10.1002/anie.202004433.
- [49] G. Nikiforidis, R. Cartwright, D. Hodgson, D. Hall, L. Berlouis, Factors affecting the performance of the Zn-Ce redox flow battery, Electrochim. Acta 140 (2014) 139–144, https://doi.org/10.1016/j.electacta.2014.04.150.
- [50] M. Krishna, E.J. Fraser, R.G.A. Wills, F.C. Walsh, Developments in soluble lead flow batteries and remaining challenges: An illustrated review, J. Energy Storage 15 (2018) 69–90, https://doi.org/10.1016/j.est.2017.10.020.