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Lithium-Free Redox Flow Batteries: Challenges and Future Prospective for Safe and Efficient Energy Storage

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Considering the costs, waste, and impact on the environment of current energy consumption, accurate, cost-effective, and safely deployed energy storage systems are required. Lithium (Li)-free redox flow batteries (RFBs) are a feasible solution. RFBs can store enormous amounts of energy effectively and are increasingly used for large-scale applications. The use of RFBs has significantly enhanced the performance of energy storage systems and effectively reduced the costs and wastage of energy storage operations. Vanadium-based RFBs are an emerging energy-storage technology being explored for large-scale deployment owing to their numerous benefits, including zero cross-contamination, scalability, flexibility, extended life cycle, and nontoxic working state. This study describes the fundamental operating principles of redox flow battery-based systems as well as the design considerations and constraints placed on each component. It discusses recent progress in the design and deployment of RFBs for energy-related applications and the remaining obstacles and prospects. Finally, this study highlights the enormous potential of RFBs and suggests some solutions to scale up the use of RFBs in the near future.

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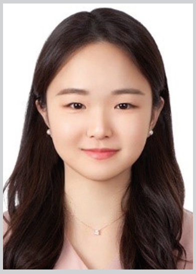
1. Introduction

Modern society is moving towards using less oil and other fossil fuels by creating electric vehicles (EVs) and producing more renewable energy; however, the need for power is steadily rising.[1–3] To meet the "Net Zero Emissions by 2050" scenario, the International Energy Agency forecasts a doubling of electricity's share in the global energy mix from 27% in 2019 to 60% in 2030.[1] Renewable energy, particularly wind and photovoltaic (PV) energy, has grown rapidly in recent years and will soon overtake all other forms of energy as a major global source. However, the unpredictable and sporadic nature of renewable energy has significantly impacted the electricity grid, potentially jeopardizing its stability.[4,5] However, developing nations continue to face serious issues as a result of the waste, expenses, and environmental impacts of such institutions. Therefore, an appropriate energy storage system is required for renewable energy power generation to stabilize intermittency and balance supply and demand.[6] The widespread use of renewable energy sources has grown in importance as a strategy for global energy security and sustainable development. Large-scale energy storage technology is a crucial core technology that makes this possible.[7,8] Energy storage methods have been applied at various scales. In 2018, pumped hydro- and compressed-air storage accounted for over 96% of the total global storage capacity.[9] Different types of batteries can be used for electrochemical energy storage, making it an excellent option for smaller storage applications because of the adaptability, high power-to-size ratio, and high efficiency. Batteries also require a smaller initial investment than pumped hydro and compressed air energy storage and do not require a particular terrain to be employed. LIBs are popular and have high energy densities; however, they are suffering from uncontrolled heat production and economically unfavorable.[10,11] A more serious issue is the gradual depletion of the global lithium supply, which limits their use. Owing to their advantages over competing technologies in terms of cost, flexibility, depth of discharge, rapid response, and safety, RFBs are useful for large-scale SES (10 kW–10 MW).[12,13] They can be used for various tasks, including load leveling, peak shaving, and emergency backup options. The power and energy modules of RFBs are created separately, have endless capacity and great security, and are simple to expand. They are an ideal option for large-scale energy storage because of their long life, high efficiency, good performance, environmental protection, and high cost performance in energy storage applications.[14–16] RFBs are a groundbreaking type of rechargeable battery specifically engineered to store electrical energy using liquid electrolyte solutions. In contrast to traditional batteries, which store energy within the electrodes, flow batteries employ external tanks to house the electrolytes. This unique configuration allows flow batteries to separate power and energy, providing scalability and versatility for a wide range of applications. At the heart of a flow battery system is an electrochemical cell containing two electrodes submerged in distinct electrolyte solutions. These solutions typically consist of redox-active species dissolved in solvents and undergo reversible electrochemical reactions during the charging and discharging processes. A selectively permeable membrane divides the electrolyte solutions within the cell, facilitating ion transfer while preventing mixing. During charging, electrical energy from an external source stimulates the electrochemical reactions, leading the electrolyte solutions to undergo oxidation and reduction. This mechanism stores energy in the form of chemical potential within the electrolytes. Conversely, discharging the battery involves releasing stored energy as electrons travel through an external circuit, ultimately producing electrical power.[17,18] An electrochemical cell and two tanks constitute the core of an RFB. Two electrodes, a separator, and an electrochemical cell constitute this device.[19,20] The pumps move the electrolyte solution between these elements. The electrolyte is known as either the catholyte or the anolyte because the redox-active cathode and anode components are dissolved in it rather than being produced as solid electrodes. The redox-active substance is impermeable to the separator but permeable to the supporting electrolyte (conducting salt). With this configuration, which is comparable to fuel cells, it is possible to expand battery power and capacity independently.[21–23] Figure 1a shows a schematic of this configuration of RBF. A related type of flow battery is hybrid flow batteries (HFBs). In HFBs, at least one redox couple with a stable redox state is observed (Figure 1b). During the charging process, the active material is electroplated on an electrode, and during the subsequent discharging process, it dissolves in the electrolyte. As electroplated metals frequently produce dendritic structures that can result in a short circuit or pierce the membrane, this cell design must be used.[24] Among RFBs, HFBs have the following benefits: mature technology, broad range of applications, low maintenance costs, high capacity for load balancing, and long cycle life. Large-scale renewable SES is currently in favor because the initial commercial operation has been accomplished.[14,19,20]

Flow batteries have several advantages. By setting the tank volume and cell stacks (reaction cells) separately, the power and capacity of the system can be scaled independently. This makes it possible to adapt precisely to connected generating units. Flow batteries are intended to store electricity for several hours, although they can transition between charging and discharging in less than a second.[25,26] Modularized flow batteries can also be relocated and set up as “mobile” energy storage systems in the shape of shipping containers.[27] With this approach, it is possible to guarantee the provision of electricity in decentralized areas, such as developing nations. Safe battery operation is ensured by using non-combustible aqueous electrolytes. Their lifespans are substantially longer than those of lead-acid and LIBs batteries. These benefits can



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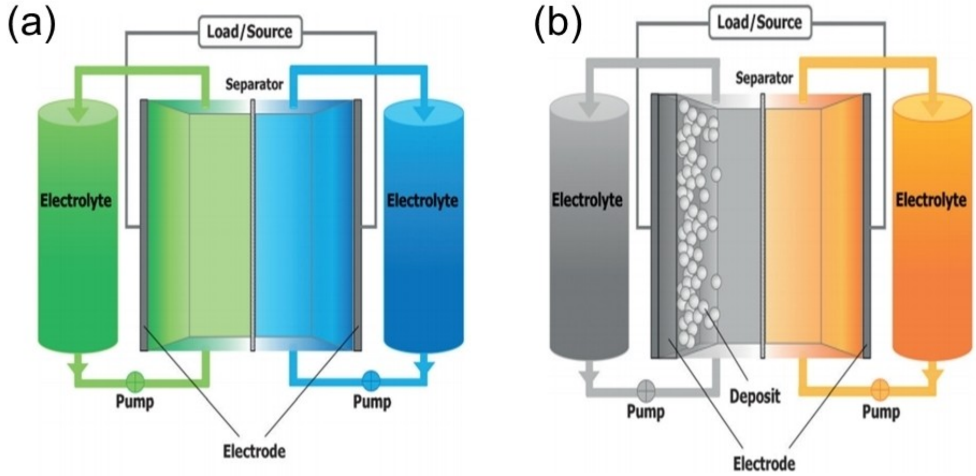

Figure 1. a) Schematic diagram of an RFB. An electrochemical cell with two compartments and a separator between them makes up the battery. Pumps move the electrolyte back and forth between the cell and the storage tanks. b) Schematic diagram of an HFB. During the charging process, a substance is electroplated onto one electrode. Reproduced with permission from.[24] Copyright 2023, Wiley-VCH GmbH.

be applied to scenarios such as peak shaving, load balancing, and frequency regulation.[28–31] Peaks in energy production may result from the rising amount of electricity produced by renewable sources such as solar and wind energy. For instance, around midday, PV electricity production tends to increase significantly but energy consumption is relatively low compared to that in the evening.

During stormy weather, wind energy can provide a significant amount of electricity.[32] An energy profile can be used to track these events. These profiles can be flattened (also known as "peak shaving") using flow batteries. Large energy-storage devices, when viewed as a full system, lessen the possibility of blackouts caused by net frequency exceedances due to overproduction.[33,34] To achieve this, massive energy-storage systems with a minimum 20-year lifespan must be built to provide reliable and affordable energy storage. These large-scale devices operate differently from small-scale proof-of-concept flow batteries. Practical applications depend on softer settings to increase the lifetime of the material. However, laboratory-scale experiments are frequently conducted under harsh conditions to explore the qualities of the material. Intelligent battery management systems and hydraulic and electric cascading of several cell stacks must be handled by large-scale systems. Owing to the requirement for the coordination and harmonization of several components and auxiliary devices, the development efforts for these complex systems are high.[35,36]

The costs of acquisition and operation are key factors that affect market penetration. In contrast, to lead and lithium, the price of vanadium as a commodity is significantly influenced by the volume of high-performance steel produced. From 2004 and 2009 saw tremendous volatility, which occurred again as China's economy expanded in the following years.[37,38] The potential cost savings of organic charge-storage RFBs compared with those made of metals is a significant advantage. Darling et al. conducted several cost evaluations that showed that the price of the active material and the cost of the membrane were the two main cost drivers of RFBs.[39–41] The most expensive separators are perfluorinated proton-exchange membranes, followed by ion-exchange membranes and porous separators. Therefore, the use of organic charge-storage compounds based on polymers is appealing.[42] Additionally, flow batteries using organic solvent-based electrolytes are not cost-competitive.[43] Therefore, from the standpoint of cost and safety, water combined with sodium chloride is suggested as the supporting electrolyte.

The prevalent flow battery systems (VRFB, iron-chromium, and zinc-bromine) exhibit plainly expressed concerns regarding the environmental or social impact during the manufacture of primary materials, damage, or disposal. For instance, mineral mining may be carried out in underdeveloped nations with subpar social and environmental regulations (such as zinc mining in China) or the electrolytes used may pose serious risks.[44,45] The control mechanisms and continuously running pumps that move electrolytes between the electrochemical cell and storage tanks further reduce the overall efficiency of flow batteries. The overall efficiency may also be affected by the active temperature control required by VRFBs.

The long-term stability of electrolytes is a considerable challenge for organic charge storage compounds. When organic compounds are oxidized or reduced, they frequently undergo side reactions, particularly if radical ions are generated. This is in contrast to metals that form stable ions. Electrolyte reactions are also observed. For instance, disproportionation reactions occur in the commonly used redox-active cathode material TEMPO at pH levels lower than 2.5 and the oxoammonium cations are unstable under basic conditions. Therefore, a long lifecycle of the active elements is only possible under ideal circumstances.

This article addresses the current advances in the creation of Li-free RFBs and their role in the creation of affordable, environmentally friendly, ample, and sustainable energy storage systems, with an emphasis on the advantages and disadvantages of such technology. After addressing the need for adequate and sustainable energy storage solutions, the distinctive features of RFBs are discussed. The methods used to prepare RFBs and noteworthy developments in their use as energy storage devices based on various electrochemical methods are outlined. Finally, we provide a summary of the present difficulties and potential future gains and offer a fresh comparison between RFBs and current energy storage technologies.

2. Li-free RFBs for Safe and Efficient Energy Storage

Over the past few years,[46] there has been an emphasis on achieving a sustainable energy transition, which involves moving away from conventional fossil fuel sources and transitioning towards renewable and regenerative sources.[47] The background is the rapidly increasing CO_2 concentration in the atmosphere which has caused unprecedented global warming. This concern has led to various national and international efforts, the most prominent of which are the Paris Agreement[48] and European Green Deal.[49] These agreements were reached to reduce CO_2 emissions in several sectors and halt global warming. Whereas the preceding decade emphasized the advancement on advancing EVs technology to enhance mobility and transportation, the upcoming decade places paramount importance on mitigating the adverse effects of industrial operations and primary energy consumption. Strides have been made towards establishing solar and wind farms as sustainable energy sources. Nonetheless, there is a drawback to the rapid expansion of renewable energy sources as the existing electric grid has not been designed or equipped to handle such a shift, resulting in a mismatch between renewable energy supply and demand. Backup facilities relying on fossil or nuclear fuels are often employed to avoid potential blackouts.[50] To guarantee a dependable electricity supply, the creation of large-scale storage facilities capable of producing power in the MW–GW range is critical. Based on the required response time, various technologies, such as compressed air storage, hydropower, flywheels, and batteries can be used.[51]

Before the early 2000s, lead-acid batteries were the primary technology used to store battery energy. These batteries are commonly used for starting, lighting, and ignition in vehicles such as boats, ships, and cars. Lead acid cells are crucial components of large-scale stationary battery systems, such as those used for uninterruptible power supplies, and they continue to be used in this market today. Although several alternative battery chemistries have been suggested and made available, niche applications such as telephone exchanges and communication hubs still rely on cells that use nickel/iron and nickel/cadmium.

Over the past few years, notable advancements have been made in battery technology, particularly in LIBs. The development of EVs worldwide, the resulting scale-up effects, and significant research efforts have led to lower costs, increased efficiency, and improved lifespans for LIB systems.[52] Although large-scale energy storage devices, such as LIBs, have potential for SES, they have limitations stemming from a range of factors, including discharge depth, thermal management, capacity fading dependent on cycle count, operational duration (i.e., how long the battery can be charged/discharged), self-discharge, and safety concerns.[53]

RFBs have several advantages over LIBs. Flow batteries were conceived in the nineteenth century and the chemistries involved have evolved. Although flow batteries were briefly employed to power the airship La France in the late 1800 s,[54] they were mostly disregarded until the 1970s. In the 1970s, notable research initiatives were launched by NASA and Japan's National Institute of Advanced Industrial Science and Technology.[55] Over the last two decades, flow batteries have grown in popularity as an energy storage technology, particularly for large-scale storage applications. Commercially available RFBs such as those using vanadium or zinc-bromine have several advantages, including minimal capacity fading during cycling, low self-discharge, and nonflammability. Additionally, these batteries offer the flexibility to independently design their power and storage capacities, enabling prolonged charge and discharge durations ranging from 4–10 hours.[56] Despite their advantages, flow batteries remain a niche technology with an installed capacity barely exceeding the GWh range. A key impediment is the relatively high installation cost of flow battery systems, which is often substantially higher than that of LIBs. Flow batteries face limited market penetration owing to their higher perceived cost compared with LIBs, as customers often prioritize the price per kWh. However, to compare the competitiveness of the different battery technologies, it is necessary to assess the economic performance of each system.

Flow batteries, also known as regenerative fuel cells, function by converting electrical energy into chemical energy during charging and then releasing it during discharging. Unlike typical batteries that store energy in active electrode materials, flow batteries accumulate energy in their liquid electrolytes. The energy produced by flow batteries depends on the electrolyte volume and concentration of the electroactive species, whereas the power generated is influenced by the electrode size and number of cells in the battery stack. A distinct characteristic of flow batteries is their ability to separate energy and power. Creating dependable, efficient, and competitive products requires a constant focus on electrochemical engineering issues.

Despite negative environmental impacts such as air pollution and climate change, the use of energy is increasing at a rapid pace. Despite increasingly clear warnings from climate scientists, the use of fossil fuels for energy consumption has persisted and increased by 2.3% increase in 2018.[57] The energy demand is rapidly rising, with estimated global annual consumption rates from 16 to 27 and 50 TW by 2050 and 3000, respectively.[58] As a result, there is a pressing need to transition towards renewable and sustainable energy sources and reassess the current levels of consumption.

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Constructing large-scale energy storage systems that effectively maintain the stability and balance between energy supply and demand is increasingly challenging owing to the intermittent and diverse nature of renewable energy sources such as solar, wind, and tidal power. In 2019, flow battery systems produced only 0.25 GW hours of stationary energy, while LIBs generated 8.8 GW hours.[59] Nevertheless, the drawbacks of LIBs, including restricted sources, inadequate recycling options, overheating, expensive production, and safety concerns,[60] have stimulated the exploration of electrochemical energy storage systems for the stationary power sector. Despite the advantages of flow batteries in terms of practicality, their high cost compared with LIBs poses a challenge to their widespread adoption in the energy storage industry.

Reducing the levelized cost of storage and capital expenditure while enhancing energy efficiency and durability remains a significant obstacle to the advancement of flow battery engineering.[61] Therefore, there is a requirement for research that encompasses both technological and scientific awareness, along with comprehensive case studies and detailed descriptions of technological scale-up. It is essential to consider the overall environmental effects, which include the indirect expenses of acquiring, reusing, and eliminating the product throughout its life cycle, from creation to disposal. This differs from focusing solely on the expense of the flow battery module or the energy or power of a single unit. Future development and applications of flow batteries should be based on conceptually sound knowledge, realistic yet ambitious research goals, and economically sound engineering.

2.1. Working Principle

A flow battery is an electrical storage device that is a hybrid of a battery and fuel cell. The core of the device consists of positive and negative components that are partitioned by a membrane through which a liquid electrolyte composed of metallic salts is pumped. Electricity is produced through ion exchange between the cathode and anode. Most commercial flow batteries utilize sulfuric acid with vanadium salt as the electrolyte and bipolar graphite plates as the electrodes. Vanadium is a limited material that can effectively prevent corrosion. Attempts have been made to incorporate precious metals such as Pt into flow batteries, similar to their use in fuel cells. The search for low-cost, widely available materials is ongoing. The flow batteries operated by pumps performed best at capacities greater than 20 kWh. According to previous reports, these batteries have an expected lifespan of approximately 20 years and can complete over 10,000 full cycles. To attain the desired voltage levels, numerous cells were interconnected in a series configuration, with each cell generating an output of 1.15–1.55 volts. The battery had a specific energy level of approximately 40 Wh/kg, which is similar to that of lead-acid batteries. The power density and acceleration rate of the flow batteries were moderate and comparable to those of fuel cells. Therefore, this battery is better suited for storing large amounts of energy than for use in electric powertrains or load leveling, which require rapid action.

Electrolyte storage tanks were used to store the electrolytes. By utilizing pre-existing storage tanks, the energy density can be enhanced by increasing the tank capacities, resulting in an estimated cost increase of only 50% compared to a completely new system. Notably, the electrolyte can be reused during battery replacement, leading to substantial cost reduction. However, these membranes pose challenges owing to their high cost and susceptibility to corrosion. Additives have been used to address this issue. Figure 2 illustrates the concept of a flow battery.

Vanadium has been extracted from countries such as China, Russia, and South Africa. Currently, 90% of the lower-grade vanadium is utilized as a steel-strengthening additive. However, battery experts, mining firms, and policymakers are optimistic about the potential of vanadium as a critical metal for renewable energy.

The expenses of a flow battery can be categorized into two primary types: power and energy. Power expenditure, which includes components such as stacks, pumps, pipes, and power electronics, can exceed $1,500/kW. On the other hand, the energy cost, which includes the costs of the tanks and electrolytes, exceeds $300/kWh. Large-scale flow batteries with capacities greater than 100 kWh have been in operation in Japan since 1996. Currently, Japan contains installations boasting capacities reaching multiple megawatts and a project is underway to develop a 60 MWh flow battery specifically designed for frequency regulation. There are trends towards reducing the cost and size of flow batteries. Instead of constructing massive single batteries that resemble chemical plants, modern solutions store stackable batteries with a typical capacity of 250 kWh. The popularity of flow batteries is increasing in Europe and began with the granting of the first patent for a titanium chloride flow battery in July 1954. In 1986, the University of New South Wales in Australia filed a patent application for a vanadium redox battery, which became a prominent technology in the field. The term "redox" derives from the electron transfer process of reduction and oxidation.

Flow batteries store electrical energy using liquid electrolytes with redox-active chemical species that flow continuously and are rechargeable electrochemical devices. Figure 2 depicts the energy conversion process in bipolar electrochemical flow reactors. These reactors employ a continuous recirculation system in which the electrolytes flow between the reactors and holding containers and are thoroughly mixed. One electrolyte contains a redox couple with a negative standard potential and the other contains a redox couple with a positive standard potential. Flow batteries, which are essentially electrochemical flow reactors, function in batch recirculation mode for energy storage and possess reversibility. The power rating of the system is determined by the quantity and size of the individual cells, whereas the energy capacity depends on the concentration of the redox species in each electrolyte and the volume of the electrolyte stored in the tanks. This particular design provides a remarkable level of flexibility in designing the power and charge capacity.

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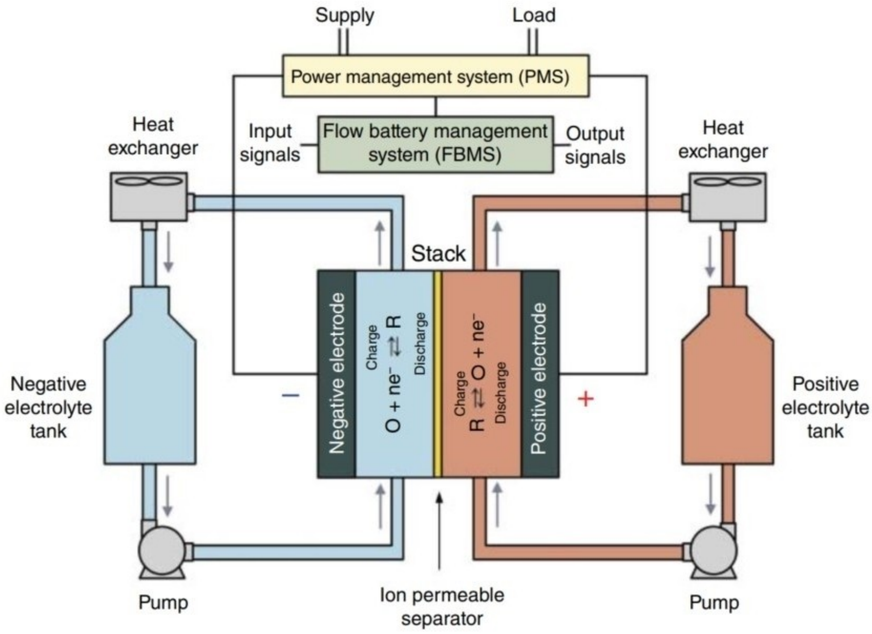

Figure 2. Flow battery system and its main components.[62] Copyright 2023, John Wiley and Sons.

During the charging process, flow battery cells operate in an electrolytic state, which consumes energy. The redox couple of the positive-electrode compartment underwent oxidation, whereas that of the negative-electrode compartment decreased. During the discharge process, reverse galvanic reactions or spontaneous electrochemical reactions occur, releasing electrical energy to an external load. Common electrode reactions include the following.[62]

$$\textit{Negative electrode } O_{-} + ze^{-} \underset{discharge}{\overset{charge}{\rightleftarrows}} R_{(-)} \tag{1}$$

$$\textit{Positive electrode } R_{(+)} \underset{discharge}{\overset{charge}{\rightleftarrows}} O_{(+)} + ze^{-} \tag{2}$$

$$\textit{Overall cell reaction } O_{(-)} + R_{(+)} \underset{discharge}{\overset{charge}{\rightleftarrows}} R_{(-)} + O_{(+)} \tag{3}$$

where z is the electron stoichiometry and O and R are the oxidized and reduced chemical species, respectively. The negative and positive redox couples are denoted by subscripts $(-)$ and $(+)$, respectively. Equation (3) represents a balanced reaction when z is equal for both the positive and negative redox couples.

2.2. Primary Components of Flow Battery System

While cell stacks are crucial components of a flow battery, the system's functioning relies on the plant balance, as well as the operations of both the Power Management System (PMS) and the Flow Battery Management System (FBMS). In the following sections, we discuss these components and outline their roles in detail.

Cell stacks: A cell stack in a flow battery is formed by interconnecting a group of identical individual cells in series and placing them on bipolar plates, with each plate featuring cathodic and anodic surfaces. The current received by each cell in the stack is identical and the voltage across the stack is the cumulative sum of the individual cell voltages. In certain designs, fused frames have been suggested, although it is more common for cells to be compressed between rigid endplates using tie bolts. Perfect hydraulic sealing must be achieved through the careful management of dimensional tolerances, component compression, and sealing gaskets. Therefore, providing a highly consistent reaction environment is desirable. In an ideal scenario, this indicates a uniform pressure drop and fluid flow, along with regulated potential and current distributions in every cell. The stacks are divided into various subcomponents.

Bipolar plates: Composite materials used in bipolar plates often contain resins, conductive additives, polymer fillers, or graphite particles. Popular materials for balancing flexibility, electrical conductivity, chemical stability, and robustness include polyvinyl alcohol, polyvinylidene fluoride, and high-density polyethylene. The plates must be simple to seal and impermeable. Their production method depends on the type of polymer filler, with injection molding or extrusion being the most common methods. HFBs can utilize planar bipolar plates as electrodes on which a metal or metal oxide is inserted. Under other circumstances, interdigitated flow fields can be added to the bipolar plates using computer numerical control machining or during the molding process.

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Separators: Separators act as tangible obstacles that separate the two electrolytes while still allowing ionic currents to pass through. Although anion-exchange membranes are being increasingly considered, perfluorinated cation-exchange membranes are the most widely used. Membranes are expensive and are a major cost in producing stacks. They have a significant impact on maintaining electrolyte balance and enabling long-term energy storage. To ensure optimal performance, ion-permeable separators have been employed to minimize the mixing of water and redox species.[63] Impurities that can precipitate onto the membrane or render it inactive should not be present in the electrolyte. Microporous separators can also be employed in certain chemistries where the two electrolytes have similar base compositions. A microporous polyethylene separator is inexpensive and suitable for precipitation impurities.

Current collectors: Current collectors are commonly made of metals such as copper and function as terminals for electrical connections. They are mechanically compressed to establish an electrical connection with the bipolar electrodes located at the ends of the stacks. The electrodes are composed of monopolar plates. To ensure a low-resistance connection and maintain the integrity of the seals, gaskets, and plates, several factors must be considered, including the stiffness of the end plates, compression levels of the polymer frame and gaskets, and dimensional tolerances. In some instances, conductive materials such as compressible graphite polymer sheets and bare or nickel-coated copper foam are employed to enhance the electrical connection and the contact pressure between the graphitized plates and the current collector.

Flow frames: Flow frames are responsible for enclosing the bipolar plates and creating a pathway for the flow of electrolytes between them and the membrane. They typically feature a porous electrode, and their thickness is influenced by the degree of applied compression. These frames incorporate several flow characteristics and are made of chemically resistant materials.[64] Grooves or channels are incorporated to facilitate the distribution of fluid flow and reduce shunt currents by creating pathways for the electrolyte to move from the manifolds into the electrode. Flow frames can utilize clip-in architectural interlocking frames, additional seals and gaskets, and clamping provisions to secure ion-permeable separators. Techniques for resin wrapping and peripheral welding such as vibration welding may be accessible.

Tanks: Tanks are composed of composite polymers or have a polymer lining, both of which must be resistant to chemicals and non-conductive. Off-the-shelf chemical tanks are less expensive but large installations require unique designs that are well supported by scaffolding made of steel with a polymer coating. Tanks can be separated into sections by filament winding or injection molding, depending on their size. To prevent the stratification or bypassing of moderately viscous electrolytes and ensure proper mixing of stored liquids, a pump bypass loop can be utilized. For maintenance purposes, tanks are frequently positioned beneath the stacks to aid gravity-assisted drainage.

Pumps: Centrifugal pumps are commonly used in flow batteries; however, positive-displacement pumps can also be utilized if their wetted parts are chemically compatible with electrolytes, nonconductive, and corrosion-resistant. Although the energy requirements for pumping might make up only a small proportion of the total energy loss in a system with a moderate pressure drop, it is still vital to prioritize energy efficiency.

Heat exchangers: Excess heat must be released into the surrounding space to maintain a controlled temperature. In most situations, heat exchangers are required, either as coils inside tanks or as components of circulation loops. Passive radiators can be utilized in chilled environments; however, air-blown coolers are more typical. Insulation or heating may be necessary to prevent flow batteries from freezing in cold climates.

Piping: Materials, together with any additional components, such as valves and joints, should be nonconductive and chemically compatible. Hoses can be used in some areas of the rest of the plant. Because electrolytes are corrosive, common electrolyte materials include fluorinated polymers and chlorinated PVC.

PMS: To meet the demands of the power grid and various applications, the AC-DC converter, referred to as the PMS, is responsible for transforming the DC generated by electrochemical cells into AC and vice versa.

FBMS: The FBMS is composed of digital hardware running a control algorithm that manages the charge-discharge regimen, regulates component operation, and shuts down the flow battery in case of an issue. It continuously monitors important parameters such as the State of Charge (SoC), voltage, current, temperature, tank level, pressure, and flow rate.

2.3. Strengths and Opportunities

According to the findings of this study, RFBs have become the dominant choice in the SES market. This can be attributed to the advantageous balance between the kinetics and thermodynamics of the half-cell electrode reactions and the water solubility of the redox species.

In addition to chromium-iron, zinc-cerium, zinc-halogen, and all-vanadium RFBs,[65–67] numerous studies have been conducted on various inorganic redox pairs with potential applications in RFBs. These alternative redox pairs have been extensively studied to explore their feasibility and suitability for use in RFB technologies. Examples of alternative inorganic redox couples that have been investigated for potential use in RFBs comprise titanium-manganese,[68–70] zinc-iron,[71] hydrogen-bromine,[72–77] polysulfide-polyiodide,[78,79] polysulfide-polybromide,[80,81] and a range of other low-cost metal-ion couples.[82,83] Despite their potential, these combinations have not surpassed all-vanadium chemistry in the SES market, mainly because of concerns related to corrosion and side reactions.

Ambipolar vinazene is considered the most viable organic pair[84] for RFB chemistry but its commercialization was discontinued after a Michigan-based company's Phase 2 ARPA-E funding expired in 2015.[84] All known organic redox couples have a common disadvantage in terms of durability. Although vanadium redox flow batteries (VRFBs) and other flow batteries exhibit better cycle and calendar lives, as well as lower production expenses than LIBs,[85] their limited energy efficiency represents the primary barrier to their extensive application in SES.[85] Porous electrodes consisting of carbon fibers with diameters ranging from 0.1–2 m and a wide distribution of pore sizes can significantly improve voltage energy efficiency and area-specific power in flow batteries. These porous electrodes also allow for a controllable pressure drop within the system, further enhancing performance. As of late 2022, VRFBs were being produced industrially but electrodes with fibers smaller than one micron were still at a low technology readiness level and were not utilized commercially.[84,86] The disregard for the limited electronic conductivity of the electrode fibers was a limitation of this study. A porous electrode can be influenced by these effects, resulting in a minimum current distribution,[86,87] which implies that most of our findings are not applicable to such scenarios.

Carbon nanofibers with significantly high electronic conductivity ranging from 650–900 $S\,m^{-1}$[88,89] can typically be produced at a carbonization temperature of 900 °C, which is approximately ten times higher than the peak conductivity observed in aqueous sulfuric acid. Another limitation is that the anisotropy of the electrode characteristics was ignored. Earlier research has shown that arranging electrode fibers parallel to the direction of the redox-fluid flow can effectively reduce the pressure drop within porous electrodes while maintaining a high area-specific power.[90] Current competition to achieve the highest peak area-specific power (over 1 W/cm^2)[91,92] in RFBs, under conditions such as low single-pass reagent utilization, 50% discharge energy efficiency, and $\leq 1\ cm^2$ cells, is pursuing misguided objectives. Although the peak area-specific power is valuable for monitoring cell design advancements, the current requirement for SES applications is to minimize the cost per unit area of the stack while achieving maximum energy efficiency during operation. It is worth emphasizing that, unlike solid electroactive material (SEAM) batteries, the optimal energy efficiency of RFBs, which is a combination of their faradaic and voltaic efficiencies, is not obtained at the lowest current density.

It should be noted that despite having longer cycle and calendar lives, flow batteries usually exhibit lower cycle energy efficiencies than batteries composed of SEAMs. The reason behind this is the internal short-circuiting current, also known as cross-over current, which generally ranges from 1–10 $mA\,cm^{-2}$. Furthermore, to mitigate the effects of crossover and minimize capital costs, RFBs typically operate at a current density approximately 100 times higher. Hence, it is crucial to strike a delicate balance between the primary advantages of RFBs, such as reduced capital cost of energy in multi-hour cycles, longer lifespan, and lower energy efficiency. Interestingly, the cost of the input energy may be a crucial factor in deciding whether to choose a flow battery, such as a VRFB, or SEAM battery, such as an LIB. If the input energy costs are lower, VRFBs may become a more attractive option, especially for applications that require frequent cycling, such as daily cycling. In the SES market, there is a pressing demand for VRFBs (as well as other durable metal-ion RFBs) to reduce the cost per unit area of the stack while ensuring that material durability is not compromised. This necessitates the development of membranes, porous electrodes, bipolar plate production processes, and materials at low manufacturing costs.

It is worth mentioning that in modern RFB designs, the ionic resistance of the electrodes is typically greater than that of the membrane. Therefore, creating less expensive (yet durable) membranes is more important than developing better conducting membranes. If carbon fiber electrodes with submicron diameters can be produced at a lower cost, the performance of RFB could be significantly affected by their use. RFB manufacturers, end users, and financiers should recognize the potential risks associated with deploying systems without reliable accelerated durability tests, especially in terms of unknown life expectancies and failure modes. Reserve (emergency) power is an industry to which RFBs are ideally suited. RFBs are expected to play a crucial role in this industry by replacing diesel generators in this application, the extended half-cycle duration (> 24 h), absence of self-discharge, safety measures, and durability (such as withstanding earthquakes and storms) take precedence over energy efficiency because charge-discharge cycles are infrequent. This industry was among the first to adopt RFB technology and offers several profitable niches, as evidenced by the success of Lockheed-Martin, Raytheon, and Ameresco. Finally, off-grid markets, such as islands, are another lucrative market for VRFBs,[93] particularly when used in conjunction with solar panels and wind turbines.

RFBs are a type of rechargeable battery technology that has received increasing attention in recent years owing to their potential applications in large-scale energy storage. Unlike conventional batteries, RFBs store energy in liquid electrolytes, which are pumped through an electrochemical cell to generate power. Electrolytes contain dissolved redox species that undergo oxidation and reduction reactions at the electrodes, allowing for energy storage and release. As research in this field continues to advance, RFBs will likely become more efficient and cost-effective, making them even more attractive energy options.

2.4. Strengths of RFBs

- **Scalability**: One key advantage of RFBs is their high scalability, which makes them suitable for large-scale energy storage applications. The battery size is determined by the size of the electrochemical cell and volume of the electrolyte container. This implies that RFBs can be designed to meet the specific energy storage requirements of a particular application.
- **Long cycle life**: RFBs have a longer cycle life than conventional batteries. This is because the electrolyte is constantly being cycled through the battery, which helps reduce the degradation of the electrodes. The cycle life of an RFB can be

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as high as 10,000 cycles, which is significantly longer than that of conventional batteries.

- **High efficiency**: RFBs have high efficiency, which means that they can convert a large percentage of stored energy into usable electrical energy. This is because the electrochemical reactions that occur in RFBs are highly reversible, which means that little energy is lost as heat during charging and discharging.
- **Safe and environmentally friendly**: RFBs are safer than conventional batteries because the electrolyte is non-flammable and non-toxic, which reduces the risks of fire and explosion. Additionally, electrolyte can be readily recycled or discarded, thereby reducing the environmental impact.

2.5. Opportunities for RFBs

- RFBs are ideal for storing renewable energy because they can store large amounts of energy over long periods. This is important because renewable energy sources such as wind and solar power are intermittent and may not always be accessible when required. By storing energy in RFBs, renewable energy sources can be made more reliable.
- Grid-scale energy storage: RFBs are well suited for grid-scale energy storage because of their scalability and long cycle life. This implies that they can be used to store excess energy during times of low demand and release it during times of high demand. This helps balance the grid and reduces the need for fossil-fuel power plants.
- Remote power systems: RFBs are ideal for remote power systems because they can store energy for long periods without requiring recharging. Thus, they can be used to power remote communities, military bases, and other off-grid applications.
- EVs charging: RFBs can be used for EV charging because they can store a large amount of energy and release it quickly, which is necessary for EV charging. Additionally, RFBs can be quickly recharged, which makes them ideal for fast-charging applications.

2.6. Comparison with LIBs

This section focuses on the contrasting suitability of flow batteries and LIBs across various segments of the SES market. Although both LIBs and RFBs were first introduced during the Oil Crisis Period of 1975–1989, their respective trajectories diverged significantly. LIBs gained widespread popularity almost immediately, whereas the publication and patenting activities of VRFBs did not increase until the 2008 Oil Crisis. Furthermore, the patent count for LIBs surpassed that of VRFBs by approximately 25 times, whereas the number of journal articles on LIBs exceeded that of VRFBs by approximately 20 times. This stark contrast emphasizes the lower market adoption and profitability of VRFBs compared to the success achieved by LIBs.[12]

Capital cost: The increase in RFB activity since 2008 can be credited to the increasing need for long-duration niche applications in the SES market, along with the associated favorable capital costs. In most countries, the current percentage of intermittent renewables in the grid generation capacity is relatively low (~10 %). Therefore, SES with half-cycle durations of two hours can effectively fulfill most of the market demand.[94] As solar panels and wind turbines contribute more than 10 % of electric power generation globally, there is a need for longer-duration SES systems (e.g., 6 h) to meet this demand.[94]

When the thickness of electrode layers exceeds approximately 50–200 μm or SEAM loadings exceed approximately 50–100 mg cm^{-2},[95–97] the area-specific resistance increases, leading to a decrease in capacity utilization. Consequently, batteries that use SEAMs and LIBs typically exhibit a low ratio between their limiting energy (at a low current density) and peak power, typically ranging from 0.2–0.6 hours[95,98] and not exceeding 2 hours.[99] The limited ionic and electronic conductivities of the layers result in the underutilization of the dense layers during charge-discharge cycles.[100] Consequently, SEAM batteries are not financially viable for applications that require half-cycle durations spanning multiple hours.

However, RFBs provide independent scaling of power (stacks) and energy (tanks). RFBs present a cost advantage over LIBs in systems requiring half-cycle durations of 4–6 hours or longer,[101] 8 hours or longer,[102] and 18 hours or longer.[103] However, for shorter timeframes,[104] this cost advantage does not exist, mainly because of the energy- and power-scale decoupling in RFBs. The precise value of the breakeven point for a half-cycle is highly dependent on the input factors.[103] In commercial practice, most LIBs used for SES installations have nominal energy-to-power ratios of approximately two hours.[105] In contrast, RFB installations are typically designed for charge-discharge cycles lasting more than four hours.[106,107]

Durability: In addition to the cost advantage, RFBs offer another benefit over SEAM batteries: their longer usable life in terms of both cycle and calendar life. Despite limited studies lasting over 5 years under operational conditions for both types of batteries, it is generally believed that VRFBs possess a significantly longer cycle and calendar life, ranging from 13–25 years, compared to LIBs, which typically have an 8-year lifespan[102,103,108,109].

The primary cause of LIB degradation is the formation of a solid electrolyte interface (SEI) at the negative electrode.[110] Because of the formation of a SEI on the negative electrode, the electrical resistance of the negative electrode increases and the number of cyclable Li^+ decreases. The growth of SEI thickness follows a square root relationship with time in the charged state, indicating that the process is governed by the limited diffusion of Li^+ through the SEI. The degradation process accelerates when the storage temperature is higher, the battery is stored at a higher SoC, and the battery is charged at a faster rate.[111] Under standard temperature conditions of 25 °C, this process causes a reduction of approximately 10 % in the cyclable charge capacity in approximately seven months, with more significant reductions occurring under more demanding

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conditions.[112] It is important to highlight that titanate anodes exhibit higher durability than graphite anodes, primarily because of the slower growth of SEI on titanate anodes. This distinction arises from the fact that graphite anodes possess more negative standard electrode potentials.

Although the deterioration of VRFBs is less understood than that of LIBs, VRFBs are usually considered more durable. Vanadium ions serve as excellent electroactive inorganic materials that offer indefinite durability and ease of recycling at the end of the lifespan of a stack. However, this ease of recycling assumes that the V_2O_5 precipitates are recovered and utilized throughout the operational life of the battery. RFBs are expected to last for at least 15,000 cycles and degrade minimally in the first 20 years.[113,114] Remixing is a simple method for reducing crossover effects in RFBs[115] and periodic rebalancing is used to resolve the charge imbalance caused by parasitic H_2 development.[116] Consequently, we do not view these problems as deterioration phenomena but rather as crucial aspects of regular RFB operation.

Energy efficiency: The inferior energy efficiency when compared with LIBs is an important but little-discussed issue. The efficiency of an energy storage system in terms of round-trip energy can be calculated by dividing the energy discharged from the system by the energy input during charging. This covers all losses, including those associated with RFB battery management systems and pumping.[117] The energy efficiency of cycles in LIBs is usually between 90% and 98%,[103,118] considering the initial formation of an SEI on the negative electrode, which contributes to Faradaic losses. In contrast, VRFBs typically have cycle energy efficiencies of 60–75%.[118–120] Systems with VRFB capacities below 1 kW typically achieve a peak cycle energy efficiency of 80% at a current density of 0.1 $A\,cm^{-2}$.[121] LIBs and RFBs can be compared across numerous fields.

- RFBs use highly conductive acidic aqueous redox fluids with an ionic conductivity of approximately 30–50 $S\,m^{-1}$,[122] which is superior to the electrolytes used in LIBs that typically employ LiPF6 solutions in alkyl carbonate with an ionic conductivity of approximately 0.25 $S\,m^{-1}$.[123] However, this advantage is insufficient to outweigh the other factors that favor LIBs.
- Despite LIBs exhibiting a higher open-circuit voltage (OCV) of 3.20 V at 50% SoC for $LiFePO_4$ batteries compared to VRFBs (1.35 V at 50% SoC),[124–126] the OCV ratio is only approximately 2.4 and is not as significant as the third factor.
- In SES applications, LIBs typically operate at low current densities of approximately 1 $mA\,cm^{-2}$.[127] On the other hand, VRFBs require higher current densities of 150–500 $mA\,cm^{-2}$[99] to minimize power stack size and address the self-discharge effects resulting from the crossover of charged species through the membrane. To provide full transparency, $LiFePO_4$ batteries can be discharged (but not charged) at 40 $mA\,cm^{-2}$,[128] which makes them highly suitable for applications that require discharge durations of less than one hour. Overall, this shows that RFBs are less energy-efficient than LIBs, despite having a substantially lower area-specific resistance. Additionally, compared to SEAM batteries, RFBs typically demonstrate a lower round-trip energy efficiency. This is primarily attributed to the requirement to operate at higher current densities in RFBs to minimize power costs and mitigate the effects of crossover through the membrane.

Other factors: Below are several general points to consider when choosing between RFBs and LIBs.

- Owing to their better energy efficiency, LIBs offer an economic advantage over RFBs in scenarios where the cost of the input electric power is high.[102]
- Compared to LIBs, VRFBs offer greater recycling value and involve lower recycling costs. In addition, they have reduced environmental impact.[129]
- The market share of LIBs in SES, whether measured in terms of energy or power, is expected to be approximately 50–100 times greater than that of RFBs by 2022.[102]
- The price per kWh of LIBs reduced at a rate of 18%/year from 1991 until 2006 and then 5% thereafter, whereas RFBs have not yet benefited from the economies of scale.[130]
- As of July 2022, the largest operating LIB installation in the world was Tesla's 182.5 MW/730 MWh system in California, whereas the largest VRFB (Dalian Rongke in PR China) was slightly smaller at 100 MW/400 MWh.[131]

3. Examples of Li-Free RFBs

RFBs are energy storage devices that use liquid electrolytes containing charged species such as metal ions or organic molecules to store and release electrical energy. One type of RFB is Li-free RFB, which does not contain lithium in its electrolyte. Here, we discuss the working principles, strengths and weaknesses, challenges, and opportunities of several RFBs.

3.1. Mechanism of VRFBs

VRFBs use liquid electrolytes containing vanadium ions to store energy. VRFBs differ from ordinary batteries in that their electrolytes are stored in a separate stack of tanks, and the size of these tanks may change to alter the power output of the battery.[132]

Although other flow batteries have been developed over the past 30 years, VRFBs are among the most desirable because they use the same material for the anolyte and catholyte, preventing contamination between the two half-cell electrolytes.[133,134] VRFBs are the most promising and economically used RFBs.[135] Figure 3 depicts a schematic of a VRFB. It primarily comprises two essential components: cell stacks and tanks. Cell stacks transform chemical and electrical energy into reversible processes and the tank holds the electrolyte.[136] In some configurations, the membrane electrode assembly and structure of the VRFB stack are comparable to those of a proton-exchange membrane fuel cell. In some instances, the materials used are comparable. Graphite or carbon are used as electrodes, graphite is used for the bipolar plates, and Nafion and perfluorinated polymers are commonly employed as ion exchange membranes.[137]

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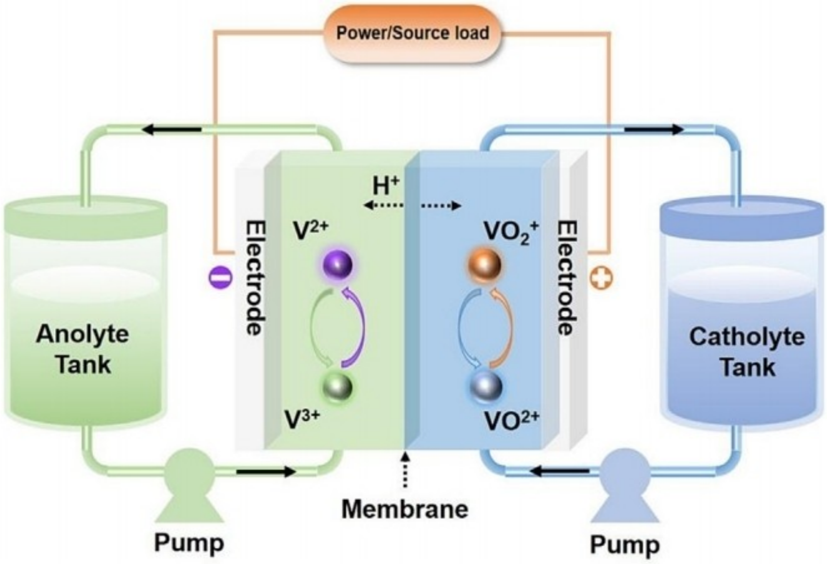

Figure 3. Graphical representation of vanadium redox flow battery.[132] Copyright 2022, Elsevier B.V.

In VRFBs, two instantaneous reactions occur at opposite membrane edges. Successful electrode reactions occur during charging and discharging.[138]

$$\text{At positive electrode}:$$
$$VO^{2+} + H_2O + e^- \leftrightarrow VO_2^+ + 2H^+ \qquad E_{cathode} = 1.004\ V \tag{4}$$
$$\text{At negative electrode}:$$
$$V^{3+} + e^- \leftrightarrow V^{2+} \qquad E_{anode} = -0.255\ V \tag{5}$$
$$\text{Overall reaction}:$$
$$VO^{2+} + V^{3+} + H_2O \leftrightarrow VO_2 + V^{2+} + 2H^+ \qquad E_{cell} = 1.259\ V \tag{6}$$

The tetravalent vanadium in VO^{2+} ions is converted to pentavalent vanadium during charging at the positive electrode, whereas trivalent ions, V^{3+} are converted to bivalent ions, V^{2+} at the negative electrode. In addition, hydrogen ions ($2H^+$) pass through the membrane to maintain the electrolyte in an electrically neutral state. VRFBs typically function at a 20–80% state of charge, meaning that the definite capacity is approximately 60% of the theoretical value. The evolution of H_2 at the negative electrode during charging and the development of oxygen at the positive electrode are recognized side reactions that may occur during operation. By moderately impeding the flow of the electrolyte and lowering the electrode surface area for the reaction, the evolved H_2 and oxygen, which take the form of bubbles, might affect the performance. In addition, the evolution of H_2 lowers the effective diffusion coefficients and the ionic and thermal conductivities compared to the evolution of oxygen.[139,140]

3.1.1. Strengths and Weaknesses

VRFBs are futuristic energy storage technologies that provide several advantages.[141–144]

- **Scalability**: VRFBs can be scaled up or down, depending on the desired energy storage capacity. Therefore, they are appropriate for various applications ranging from modest home systems to massive utility-scale installations.[145]
- **Extended lifespan**: Compared to other battery technologies, VRFBs have a cycle life of up to 20,000 cycles, which is very long.[113]
- **High energy efficiency**: VRFBs have a round-trip efficiency of about 70–80%, making them very effective. This indicates that batteries can store and release energy with minimal energy losses.[146]
- **Safe and ecologically friendly**: Compared to other battery technologies, VRFBs are safer because the electrolytes they utilize are nontoxic and non-flammable. VRFBs are environmentally favorable alternatives because of the recyclable nature of vanadium electrolytes.[63]
- **Reliable performance**: Over time, the VRFBs retain their performance with minimal deterioration or capacity loss. Consequently, batteries can deliver a constant level of performance over the course of their lifetime.[147]

Overall, because of their scalability, longevity, high energy efficiency, safety, and environmental friendliness, VRFBs are promising energy storage technologies. However, their high prices remain a barrier to their extensive commercial adoption.

Although VRFBs have several advantages, they have drawbacks that must be considered.

- **High price**: VRFBs are currently more expensive than other battery technologies, which may prevent widespread use.[144]
- **Huge size**: As VRFBs require separate tanks and membranes, their equipment needs to be larger and more complicated

than that of conventional batteries. They may be less suitable for certain applications when space is available.[142]

- **Energy density**: Compared with other battery technologies, VRFBs have a lower energy density, requiring more room to store the same amount of energy.[144]
- **Complicated upkeep**: To maintain the proper electrolyte concentration and membrane integrity, VRFBs require routine maintenance. Consequently, the overall cost and complexity of the system can increase as a result.[144]
- **Restricted temperature range**: The performance of VRFBs may be constrained under conditions of severe heat, which may reduce their effectiveness and shorten their lifespan.[144]

Although VRFBs provide several benefits, their high price and complexity may prevent their mainstream use. These issues are being addressed and efforts are being made to increase the overall performance and cost-effectiveness of VRFBs.

3.1.2. Challenges and Opportunities

For widespread implementation, VRFBs must overcome several obstacles:[144,148]

- **Price**: VRFBs are currently expensive because vanadium is expensive, and the battery is intricately designed. The price of VRFBs has been lowered by increased production and innovative manufacturing processes.
- **Energy density**: VRFBs have a lower energy density and require more room to store the same amount of energy. To make VRFBs more competitive, researchers have attempted to increase their energy density.[146]
- **High-temperature performance**: VRFBs' high-temperature performance may be compromised, limiting their usefulness in specific applications. New electrolytes and membranes that function at higher temperatures are being developed through continuing research.[149–151]
- **Efficiency**: Although VRFBs have a high efficiency level, there is potential for innovation to boost their performance and reduce energy losses during charge/discharge cycles.[152]
- **Recycling**: Although the electrolyte used in VRFBs can be recycled, doing so can be costly and energy intensive. The development of more effective and affordable VRFBs recycling techniques is currently being investigated.[153]

Ultimately, although VRFBs have several advantages over other battery technologies, major problems still need to be solved for them to become more affordable and competitive. These problems, as well as the general performance and cost-effectiveness of VRFBs, are being addressed through ongoing research and development. Many options for energy storage and integration of renewable energy have been presented using VRFBs.

- **Integration of renewable energy**: VRFBs can store additional renewable energy produced by wind and solar power, which can subsequently be used when energy production is low, or demand is high.[143]
- **Grid stability**: By storing energy during periods of low demand and releasing it during periods of high demand, VRFBs can help preserve grid stability and reduce the need for peaker plants to burn fossil fuels.[141]
- **Microgrid applications**: In areas with erratic or nonexistent grid connections, VRFBs can be employed in microgrid applications to offer energy storage and backup power.[142]
- **Electric car charging**: VRFBs are a dependable form of energy storage that can be utilized in EVs charging stations to ensure speedy and effective charging.[144]
- **Offshore energy storage**: The use of VRFBs for offshore energy storage can help overcome the difficulties posed by long-distance energy transmission.[144]

Overall, as the world moves towards more sustainable energy systems, VRFBs provide numerous opportunities for energy storage and renewable energy integration. To facilitate the wider use of VRFBs, ongoing research and development activities are focused on enhancing their performance, cost-effectiveness, and scalability.

3.2. Mechanism of Zinc-Bromine RFBs

Rechargeable batteries, called zinc-bromine flow batteries, use a zinc bromide solution as electrolyte. The battery consists of two tanks, each containing a zinc bromide solution (Figure 4). A membrane that separates the solutions allows ions to move through while preventing the two solutions from mixing.[154]

Zinc is deposited on the negative electrode, which is often composed of graphite or carbon felt, as the battery is charged. At the positive electrode, which is typically made of a porous metal such as stainless steel, bromine is created simultaneously. Zinc at the negative electrode combines with bromine at the positive electrode to generate zinc bromide, which releases electricity when the battery is depleted.[155,156] Subsequently, the zinc bromide solution is pumped back into the tanks for refilling. The electrochemical equation can be written as:[150]

$$\begin{aligned}&\textit{At negative electrode}:\\&Zn^{2+}+2e^{-}\leftrightarrow Zn\qquad E_{cathode}=-0.76\ V\end{aligned}\tag{7}$$

$$\begin{aligned}&\textit{At positive electrode}:\\&2Br^{-}\leftrightarrow Br_2+2e^{-}\qquad E_{anode}=+1.07\ V\end{aligned}\tag{8}$$

$$\begin{aligned}&\textit{Overall cell reaction}:\\&ZnBr\leftrightarrow Zn+Br_2\qquad E_{cell}=+1.83\ V\end{aligned}\tag{9}$$

One benefit of zinc-bromine flow batteries is their capacity for large-scale energy storage, which makes them suitable for applications such as grid-scale energy storage. They can also be recharged and discharged repeatedly without degradation, are reasonably inexpensive, and have a long-life cycle Routine upkeep and monitoring are necessary to ensure that batteries operate effectively.[157]

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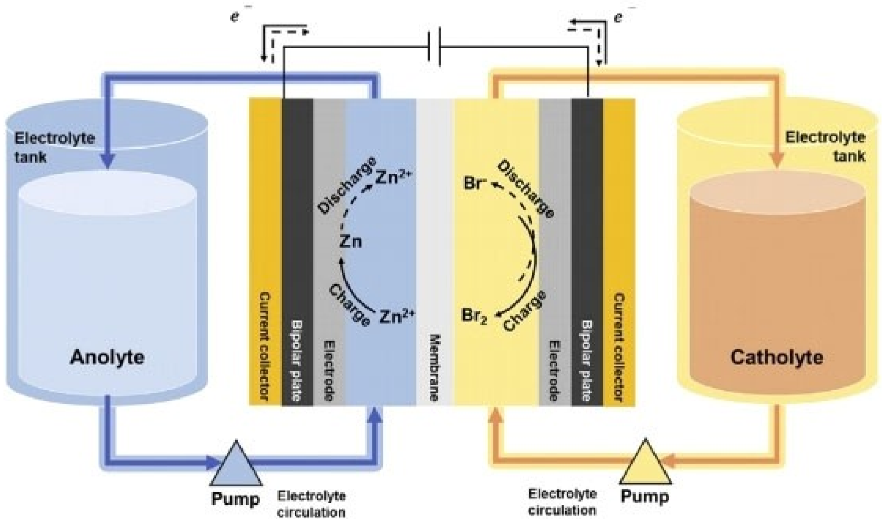

Figure 4. Schematic representation of a zinc-bromine redox flow battery.[155] Copyright 2021, Elsevier B.V.

3.2.1. Strengths and Weaknesses

The following are some advantages of zinc-bromine flow batteries:

- **Energy Density**: Zinc-bromine flow batteries can store large amounts of energy in a relatively compact space owing to their high energy density. Therefore, they are appropriate for applications requiring considerable energy storage over a small area.[158]
- **Cycle Life**: Zinc-bromine flow batteries can be charged and drained numerous times before deteriorating. Therefore, they are more affordable than other battery types that need to be replaced more regularly.[159]
- **Economical**: Zinc-bromine flow batteries are inexpensive and are thus attractive alternatives for large-scale energy storage projects.[158]
- **Environmentally Friendly**: Zinc-bromine flow batteries employ non-toxic components and do not emit any hazardous waste. Thus, they are regarded as safe and environmentally friendly.[157]
- **Efficiency**: The high efficiency of zinc-bromine flow batteries allows them to convert a significant portion of stored energy into useful electricity.[159]
- **Scalable**: To accommodate the varying energy storage requirements of various applications, Zinc-bromine flow batteries can be flexibly scaled up or down to accommodate the varying energy storage requirements of various applications. This qualifies them for a variety of uses, including grid-scale and home energy storage.[160]

The following are some zinc-bromine flow battery drawbacks or restrictions:

- **Power Density**: Compared to other battery types, zinc-bromine flow batteries have a comparatively low power density, suggesting that they might not be appropriate for applications requiring a high-power output.[159]
- **Temperature Sensitivity**: Zinc-bromine flow batteries are susceptible to temperature variations, which may impair their performance and shorten their lifespan. High temperatures can harm batteries by causing the electrolyte solution to freeze or evaporate.[161]
- **Maintenance Requirements**: To ensure that zinc-bromine flow batteries function effectively, they must be regularly maintained and observed. Maintenance includes monitoring the charge level, testing electrolyte concentrations, and replacing any broken or worn parts.[161]
- **Complexity**: Zinc-bromine flow batteries can be more challenging to design, produce, and install than other battery types because they are more complex. Thus, complex control systems are required to ensure optimal performance and control electrolyte flow.[162]
- **Corrosion**: Zinc-bromine flow batteries are prone to corrosion, particularly if the electrolyte is not carefully controlled. The battery components can be harmed by corrosion, which shortens the lifespan of the battery.[163]

3.2.2. Challenges and Opportunities

The following are some of the difficulties that zinc-bromine flow batteries face:

- **Cost**: Although zinc-bromine flow batteries are typically considered less expensive than other battery types, they require a sizable upfront investment. Wider acceptance may be hampered by the cost of components, manufacturing, and installation.[164]
- **Performance Degradation**: With time, zinc-bromine flow batteries exhibit performance losses similar to all battery technologies. Several factors, including temperature changes, overcharging, and electrolyte aging, may have contributed to this. Producers and researchers face significant problems extending the life of these batteries.[161]
- **Scale-up**: Although zinc-bromine flow batteries are scalable, they can be difficult to scale up to satisfy the requirements of large-scale applications, such as grid-scale energy storage.

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This level of production and installation requires significant infrastructure and logistics expenditure.[160]

- **Safety**: Although zinc-bromine flow batteries are typically regarded as secure and environmentally friendly, there is always a chance of mishaps or malfunctions. Strict adherence to production regulations and procedures, as well as continuous monitoring and maintenance, are needed to guarantee the safety of these batteries.[162]
- **Restricted Market**: Compared to more established battery technologies, such as LIBs, zinc-bromine flow batteries are still a relatively new technology and have a small market. Manufacturers may find it challenging to achieve economies of scale and cut costs as a result.[165]

Opportunities for zinc-bromine flow batteries include:

- **Energy Storage**: As the utilization of renewable energy sources increases, so does the necessity of energy storage. Particularly in applications that require long-duration storage, zinc-bromine flow batteries have the potential to become a crucial technology for energy storage.[159]
- **Grid-Scale Applications**: Zinc-bromine flow batteries are well suited for grid-scale applications such as peak shaving, load shifting, and frequency regulation. Its ability to store and discharge large amounts of energy over an extended period makes it a valuable asset for grid operators.[164]
- **Remote Locations**: Zinc-bromine flow batteries are an affordable option for energy storage in off-the-grid villages, military bases, and mining operations. With the help of these batteries, a dependable and long-lasting energy source can be produced without the expense of costly infrastructure.[166]
- **Research and Development**: Zinc-bromine flow batteries are a relatively new technology, and there is still much to learn about them. The potential of this technology can be realized through research and development projects focused on enhancing performance, lowering prices, and broadening applications.[165]

3.3. Mechanism of Iron-Chromium RFBs

Rechargeable flow batteries, called iron-chromium flow batteries (ICFBs), employ redox interactions between iron and chromium ions to store and release energy.[167] The basic operation of an ICFB entails the passage of charged electrolytes (ions) between two tanks or chambers. Iron ions are oxidized in one tank using a positive electrode, called an anolyte, to produce Fe(III) ions, whereas chromium ions are reduced in the other tank using a negative electrode, called a catholyte, to produce Cr(II) ions,[55] as shown in Figure 5.

The electrolytes are pushed from their tanks during discharge through a cell stack, where they are engaged in a redox reaction to generate energy. During discharge, the electrolytes are pumped from their respective tanks through a cell stack, where they undergo a redox reaction to produce electricity. The Fe(III) ions in the anolyte transfer electrons to the electrode and are reduced to Fe(II) ions. Similarly, the Cr(II) ions in the catholyte accept electrons from the electrode and are oxidized to Cr(III) ions. This process is reversed during charging. Electricity is used to oxidize Fe(II) ions back to Fe(III) ions in the anolyte and to reduce Cr(III) ions back to Cr(II) ions in the catholyte.[168,169] The following electrochemical processes enable ICFBs to store and release electrical energy.[170]

$$\begin{aligned} &\textit{At positive electrode}: \\ &Fe^{3+} + e^- \leftrightarrow Fe^{2+} \quad E_{cathode} = +0.77\ V \end{aligned} \tag{10}$$

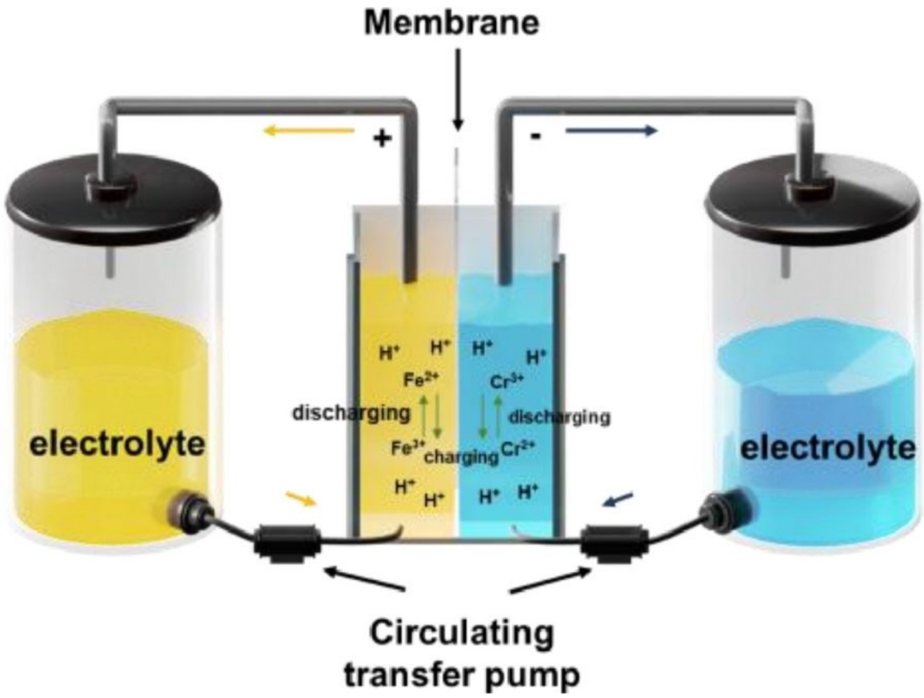

Figure 5. Schematic of Iron chromium redox flow battery.[172] Copyright 2023, Elsevier B.V.

$$\begin{aligned}&\textit{At negative electrode}:\\&Cr^{2+} \leftrightarrow Cr^{3+} + e^- \qquad E_{anode} = -0.41\ V\end{aligned} \tag{11}$$

$$\begin{aligned}&\textit{Overall reaction}:\\&Fe^{3+} + Cr^{2+} \leftrightarrow Fe^{2+} + Cr^{3+} \quad E_{cell} = 1.18\ V\end{aligned} \tag{12}$$

The size of the cell stack, electrolyte concentration, and velocity of the electrolyte flow through the system all affect the amount of energy that an ICFB can store. As ICFBs can be scaled up to provide large-scale energy storage for grid-level applications, they have advantages over other forms of batteries.[171]

3.3.1. Strengths and Weaknesses

Compared to other battery types, ICFBs have the following advantages:

- **Large Electrolyte Tanks**: ICFBs have numerous electrolyte tanks that contribute to their high energy-storage capacity. These are the best choices for grid-scale energy storage applications.[173]
- **Lifespan**: ICFBs have longer lifespans than other battery types because they do not experience the same electrode deterioration as conventional batteries. Therefore, they can function for many years before being replaced.[174]
- **Scalability**: ICFBs have a high degree of scalability, making it possible to quickly modify them to fit a variety of applications, from small-scale systems to massive industrial and grid-level energy storage systems.[174]
- **Efficiency**: During charging and discharging, ICFBs experience minimal energy loss and high energy-conversion efficiency.[173]
- **Environmentally Friendly**: ICFBs use abundant and harmless components that are easily recyclable, such as iron and chromium, making them more environmentally friendly compared to zinc-bromine RFB.[173]

Overall, because of their high energy storage capacity, extended lifespan, scalability, efficiency, and sustainability, ICFBs are viable alternatives for large-scale energy storage applications.

There are a few drawbacks to ICFBs that prevent their extensive use in energy storage applications. These shortcomings include the following:

- **Energy density**: ICFBs have low energy density, which means that a given amount of energy must be stored over a large area.[170]
- **Efficiency**: ICFBs often have lower efficiencies than other types of flow batteries, making them less economical.[164]
- **Lifespan**: ICFBs typically have a shorter lifespan than other battery types, which may result in greater maintenance and replacement expenses over time.[175]
- **Environmental issues**: Both the manufacture of chemicals used in ICFBs and their disposal of used batteries can have adverse effects on the environment.[170]
- **Corrosion**: The corrosive electrolyte of ICFBs may cause problems with the deterioration of the cell components over time.[176]

3.3.2. Challenges and Opportunities

Several obstacles prevent the widespread use of ICFBs as large-scale energy-storage solutions. The principal obstacles are as follows:

- **Price**: ICFBs are more expensive than other types of flow batteries because of the comparatively high cost of iron and chromium.[170]
- **Energy density**: Compared with other battery types, ICFBs have a lower energy density, requiring a larger physical space to store the same amount of energy.[170]
- **Efficiency**: ICFBs lose a large amount of energy during charging and discharging operations because of their low energy efficiency.[170]
- **Scalability**: Because ICFBs are difficult to scale up, it is challenging to employ them in large-scale energy storage applications.[174]
- **Lifespan**: ICFBs have a shorter lifespan than other battery types, which over time may lead to increased maintenance and replacement expenditures.[174]
- **Environmental impact**: Some users may be concerned about the environmental effects of the chemicals used in ICFBs during production and disposal.[170]
- **Technology maturity**: ICFBs are a relatively new technology; as a result, they have not been as thoroughly tested and validated as other types of batteries. This makes it challenging to obtain funding and attract customers.[174]

ICFBs are a promising technology for large-scale energy storage because they offer several advantages. The opportunities include the following:

- **Energy storage**: ICFBs are ideal for situations where energy needs to be stored for several hours or days because they can retain energy for lengthy periods.[173]
- **Flexibility**: ICFBs are versatile and can be utilized in a variety of applications, including grid-scale energy storage, backup power, and renewable energy integration.[170]
- **Safety**: Compared to other battery types, ICFBs are considered safe because they do not emit hazardous gases or pose a fire risk.[170]
- **Sustainability**: ICFBs are more environmentally friendly than other types of batteries because the materials they employ are reasonably common and simple to obtain.[173]
- **Maintenance**: ICFBs require little upkeep and can function for many years without major repairs or replacements.[170]
- **Cost-reduction potential**: ICFBs may become more affordable and competitive with other battery types because the prices of iron and chromium are predicted to fall in the future.[170]
- **Efficiency**: The efficiency of iron–chromium flow batteries has been improved by researchers, which could eventually result in lower costs and higher performance.[173]

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3.4. State-of-the-Art Uses of RFBs

Flow batteries are rechargeable batteries that utilize two different electrolytes that are stored in external tanks and flow through a cell to generate electricity. Several companies and industries use different types of flow batteries. Energy Storage Systems (ESS) Inc. provides energy storage options based on the flow battery technology. Iron flow battery (IFB) systems have been developed and marketed by ESS for a variety of energy storage applications. With an emphasis on applications such as renewable energy integration, microgrids, and commercial and industrial energy storage, ESS IFB systems are designed to offer long-term energy storage. The IFB systems offered by the company are scalable, making it simple to adapt to the particular requirements of various applications. IFB systems have been implemented by ESS for various clients, including utilities, business and industrial buildings, and military locations. The business has also worked on developing and researching other flow battery systems, including zinc-bromine batteries. Overall, ESS provides a variety of energy storage options based on flow battery technology, with the technology chosen depending on particular project requirements and client preferences.[177–179] A VRFB developed and marketed by a Japanese firm called Sumitomo Electric industries is used in energy storage applications. The business deployed VRFB systems for a range of uses, including the integration of renewable energy sources, peak shaving, and backup power. In addition to developing VRFBs, Sumitomo Electric is developing iron-chromium, zinc-bromine, and other types of flow batteries. The company's flow battery is used in grid-scale energy storage projects and is intended to provide long-term energy storage.[180–182] Uni Energy Technologies (UET) is one of the top suppliers and manufacturers of flow battery systems. VRFB technologies have been developed and commercialized by businesses for a range of energy storage applications. The VRFB systems from UET are intended to provide long-duration energy storage, which can be utilized for tasks such as peak shaving, backup power, and the integration of renewable energy sources. The VRFB systems are flexible and adaptable to the requirements of various applications. UET has implemented VRFB systems for a range of clients, including utilities, business, and industrial facilities, and microgrids. The business has also developed other flow battery systems, including ICFBs.[183–185] Redflow specializes in the development and commercialization of zinc-bromine flow battery systems for energy storage applications. Redflow's flow battery systems are designed to provide high-capacity, long-duration energy storage with a focus on applications such as residential and commercial energy storage, off-grid power, and telecommunications backup power. The company's flow battery systems are modular, allowing easy scalability and customization to meet the needs of different applications. Redflow has implemented flow battery systems for clients worldwide, including in Europe, Africa, and Australia. The business has also worked on developing and researching other flow battery systems, including ICFBs.[186,187] Flow batteries have been employed in projects by Renewable Energy Systems (RES). RES provides energy storage solutions for integrating renewable energy sources. LIBs, lead-acid batteries, and flow batteries are some of the energy storage technologies used by RES. The company has utilized flow batteries for applications such as microgrids, grid stabilization, and off-grid power systems. To supply clients with flow battery-based energy storage solutions, RES has worked with various flow battery technology providers. For instance, in 2017, RES announced a collaboration with Avalon Battery, a producer of VRFBs, to provide energy storage options for business and industrial clients. Overall, RES provides a variety of energy storage options, with the technology chosen depending on project specifications and customer preferences.[188]

4. Conclusions and Perspectives

In this review, we comprehensively outline the benefits of Li-free redox flow batteries (RFBs), emphasizing their zero cross-contamination, scalability, flexibility, extended life cycle, and non-toxicity. We also delve into prevalent flow batteries systems like vanadium, iron-chromium, and zinc-bromine, providing a detailed overview of their applications and potential. Our perspective emphasizes a multifaceted approach to unlocking the full potential of RFBs and other flow battery technologies while addressing current challenges. This approach necessitates extensive research, innovation, collaboration, and strategic planning to overcome hurdles and accelerate their adoption in mainstream energy storage solutions. Despite significant progress, achieving RFBs with efficiency and energy densities comparable to lithium-ion batteries (LIBs) remains a hurdle, demanding further efforts. Challenges include the large system size, corrosion issues, and higher costs per unit electricity compared to LIBs. However, intensive research and development efforts hold the promise of enhancing efficiency and driving long-term cost reduction in flow battery systems. Environmental sustainability is a key consideration in our perspective with vanadium, zinc-bromine, and iron-chromium RFBs offering inherent advantages due to their non-toxic and recyclable components. By positioning flow batteries as pivotal components of a sustainable energy infrastructure, collaboration among stakeholders can be fostered to accelerate their adoption and address environmental challenges. Looking ahead, our vision involves the widespread adoption of vanadium, zinc-bromine, and iron-chromium RFBs as mainstream energy storage solutions. This adoption can facilitate the integration of renewable energy sources, enhance grid stability, and expand energy access globally. Through a supportive ecosystem of research, innovation, and collaboration, this vision can be realized, paving the way for a more resilient and sustainable energy future for generations to come.

Nomenclature

C Charging
C Volumetric capacity
D Discharging

m Mass
n Number of electrons
F Faraday
Qs Constant
Q Charge
M Molar mass
V Volume
E Energy density
E Potential
T Charging time
U Voltage

Symbol

η Efficiency

Abbreviation

FBMS Flow Battery Management System
ICFB Iron-chromium flow battery
LIBs Lithium-ion batteries
PMS Power Management System
RFBs Redox flow batteries
SEAM Solid electroactive material
SES Stationary energy storage
VRFB Vanadium redox flow battery

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Conflict of Interests

The authors declare no conflict of interest.

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