

Internal Architecture and Electrochemical Mechanisms of Redox Flow Batteries

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Abstract: Redox Flow Batteries (RFBs) have emerged as a transformative solution for large-scale, long-duration energy storage, addressing the safety and scalability limitations of conventional lithium-ion systems. This paper provides a comprehensive analysis of the internal architecture and electrochemical mechanisms that define RFB performance. By decoupling power and energy capacity through external electrolyte circulation, RFBs offer unique flexibility and an exceptional cycle life exceeding 15,000 cycles. We detail the critical role of the cell stack components, including bipolar plates, carbon-based electrodes, and optimized flow field designs, in minimizing concentration polarization and enhancing mass transport. A significant portion of the study focuses on the separator as the primary gatekeeper of efficiency. We evaluate the technical pathways of membrane materials, ranging from industry-standard perfluorinated sulfonic acid (PFSA) membranes to emerging porous separators and Polymers of Intrinsic Microporosity (PIMs). These materials are assessed based on their ion exchange capacity, area resistance, and ability to mitigate active species crossover. ly, the paper reviews the industrial landscape, highlighting large-scale demonstration projects and the ongoing transition toward low-cost, non-fluorinated materials. By integrating material innovation with system-level optimization, RFBs are poised to serve as the backbone of resilient, stable, and sustainable energy grids.

Keywords: Redox Flow Batteries (RFBs); Long-Duration Energy Storage; Ion Exchange Membranes; Reaction Mechanisms; Vanadium Redox Flow Battery (VRFB)

1. Introduction

The global transition toward renewable energy systems has intensified the demand for efficient energy storage solutions capable of stabilizing the power grid against the intermittent nature of wind and solar power. While lithium-ion batteries currently maintain a dominant market position due to their established infrastructure, they are increasingly scrutinized for their inherent limitations, particularly regarding safety and scalability [1,2]. The risk of thermal runaway and explosion remains a critical concern for large-scale deployments. Consequently, the energy sector is witnessing a shift toward next-generation technologies. Solid-state batteries have emerged as a significant contender, offering higher energy density and improved safety by mitigating combustion risks, although they currently remain in the semi-solid research and development phase as they work to overcome rate performance challenges [3]. Similarly, aqueous ion batteries provide an inherently safe, non-flammable alternative due to their water-based media, yet their narrow voltage window, typically between 1 and 2 volts, results in a low energy density that makes them unsuitable for mobile power applications [4].

In this landscape of evolving technologies, Redox Flow Batteries (RFBs) represent a transformative approach specifically tailored for large-scale, long-duration energy storage. The fundamental architectural distinction of the flow battery lies in the separation of the electrolyte from the cell stack [5]. In these systems, the active materials are dissolved in

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liquid electrolytes stored in external tanks and circulated through the stack via a pumping system. This unique configuration allows for the complete decoupling of power and energy capacity; the power output is determined by the size of the electrode stack, while the energy capacity is limited only by the volume of the electrolyte tanks [6]. This flexibility allows for theoretical capacity expansion without the need for proportional increases in cell size, making RFBs the most viable solution for storage requirements exceeding four hours.

Beyond scalability, the safety and longevity of flow batteries provide a decisive advantage over traditional closed-cell chemistries. Because many mainstream RFBs, such as the All-Vanadium Flow Battery, utilize aqueous electrolytes, they are inherently non-flammable and eliminate the risk of thermal runaway [7]. This makes them ideal for massive, hundred-megawatt-level storage projects where safety is paramount. Furthermore, while lithium-ion batteries typically offer a lifespan of only a few thousand cycles, flow batteries are capable of reaching between 15,000 and 20,000 cycles [8]. Although they face challenges such as lower energy density and specific temperature requirements, ideally maintaining an operating window between 25C and 50C to prevent precipitation, their ability to provide stable, long-term service makes them the cornerstone of the impending energy system revolution.

2. Working Principles and System Architecture

2.1 Basic Components of Redox Flow Batteries*

Redox flow batteries (RFBs) feature a unique architecture that decouples the energy storage medium from the power generation component. Unlike conventional sealed-cell batteries, RFBs store energy in liquid electrolytes within external tanks [9]. Energy conversion occurs in the electrochemical cell stack, designed to facilitate efficient ion exchange and electron transfer while physically separating the positive and negative electrolyte streams.

The stack's mechanical integrity is maintained by high-strength end plates that provide the compression necessary to prevent leakage and minimize contact resistance. Internally, the stack consists of repeating current frames and bipolar plates [10]. The frames house the electrodes and direct electrolyte flow into the reaction zone. Bipolar plates serve a dual role: they act as a physical barrier between adjacent cells and provide a conductive path for series-connected electron flow. Typically made of carbon-based composites, these plates must resist corrosion; precise alignment is vital to avoid uneven current distribution or internal shunts.

The RFB's electrochemical performance is primarily dictated by the electrodes and the electrolyte. Electrodes serve as redox reaction sites and are usually composed of carbon cloth, paper, or felt due to their high conductivity, surface area, and stability. These substrates are often treated to enhance electrochemical activity, resulting in faster kinetics and higher power densities.

The electrolyte system determines the battery's energy capacity and chemical characteristics. In All-Vanadium Redox Flow Batteries (VRFBs), active vanadium ions are dissolved in an aqueous solvent, which influences the voltage window and system stability [11]. While vanadium-based systems are mature, current research explores organic molecules and halogens to improve energy density or reduce costs. The functions of these core materials and mechanical components are summarized in Table 1.

Table 1. Key Components and Functional Specifications of Flow Battery Systems

Component	Material Type	Primary Function
End Plates	Structural Alloys/Polymers	Provide mechanical compression and structural support.

Bipolar Plates	Carbon/Polymer Composites	Facilitate electron transfer and separate individual cells.
Electrodes	Carbon Cloth/Paper/Felt	Provide high-surface-area sites for redox reactions.
Separator (Membrane)	Ion Exchange Membrane (e.g., Nafion)	Prevent electrode contact while allowing selective ion transport.
Electrolyte	Active Aqueous Species	Store chemical energy and transport ions through circulation.

2.2 System Design and Optimization*

Scaling from laboratory cells to megawatt-level industrial systems requires significant design optimizations focused on mass transport efficiency and environmental control. These enhancements are critical for maintaining chemical stability and maximizing energy output across large-scale deployments.

A primary optimization focus is the flow field design etched onto bipolar plates. Electrolyte distribution uniformity directly influences Coulombic Efficiency (CE) and area capacity [12]. Without effective channels, electrolytes fail to penetrate porous electrodes, creating "dead zones" that deplete active species. Serpentine and interdigitated designs are the two primary configurations used to address this. Serpentine channels utilize a continuous winding path to create a high-pressure gradient, while interdigitated designs employ a "dead-end" strategy that forces the electrolyte through the electrode material. Both designs are essential for minimizing concentration polarization, where reaction rates are limited by reactant supply, ensuring the entire electrode volume is utilized to maximize power output.

The system's operational stability is sustained by the "Balance of Plant" (BoP), primarily the circulation pumps and thermal management systems. Pumps must move electrolytes at rates matching the current density, though they introduce parasitic power losses that must be minimized to preserve overall energy efficiency.

Thermal management is vital for long-term reliability, as the optimal operating window is narrow, typically between 25C and 50C. In vanadium systems, exceeding 45C can cause vanadium pentoxide precipitation, which clogs flow channels and damages the stack. Conversely, temperatures below 0C reduce solubility, leading to similar mechanical risks. Integrated air or liquid cooling systems maintain a safe 10-40C buffer. Additionally, monitoring the State of Charge (SOC) within a 20-80% range and managing pipe pressure differentials are essential practices to ensure the system achieves its 15,000-cycle target lifespan.

3. Performance Indicators and Polarization Mechanisms

3.1 Performance Evaluation Framework*

The viability of a redox flow battery (RFB) for commercial energy storage is determined by a rigorous set of performance metrics. These indicators quantify the efficiency of energy conversion, the capacity for energy storage, and the long-term durability of the system under continuous cycling [13].

The efficiency of an RFB is primarily characterized by three interrelated metrics: Coulombic Efficiency (CE), Voltage Efficiency (VE), and Energy Efficiency (EE). Coulombic Efficiency represents the ratio of the total charge extracted during discharge to the total charge injected during charging. High CE, often approaching 100% in mature systems like all-vanadium flow batteries, indicates minimal side reactions and low active material crossover. Voltage Efficiency, on the other hand, reflects the losses due to internal resistance and overpotentials; it is the ratio of the average discharge voltage to the average charge voltage.

The most critical benchmark for system performance is the Energy Efficiency (EE), which is the product of the two aforementioned metrics ($EE = CE \times VE$). According to

national standards mentioned in the document, commercial-grade vanadium flow batteries are expected to achieve an energy efficiency of over 80%. These efficiency parameters are summarized in Table 2 to highlight their impact on system performance.

Table 2. Key Efficiency Indicators for Flow Battery Performance

Metric	Definition	Primary Influencing Factors
Coulombic Efficiency (CE)	Q_{dis}/Q_{ch}	Ion crossover, side reactions, shunt currents.
Voltage Efficiency (VE)	V_{dis}/V_{ch}	Ohmic resistance, reaction kinetics, mass transport.
Energy Efficiency (EE)	$CE \times VE$	Overall system design and material selection.

Beyond efficiency, the performance evaluation also focuses on area capacity and cyclic stability. Area capacity refers to the amount of charge stored per unit area of the electrode, which is directly linked to the concentration of active species in the electrolyte and the uniformity of the flow field [14]. Maintaining a high area capacity is essential for reducing the physical footprint of the stack. Cyclic stability is the measure of the battery's ability to maintain its performance over thousands of cycles. Flow batteries are specifically designed for longevity, targeting 15,000 to 20,000 cycles, provided that the state-of-charge (SOC) is monitored within the optimal 20% to 80% range to avoid irreversible chemical degradation.

3.2 Losses and Technical Challenges*

Despite their advantages, RFBs experience various energy losses during operation, primarily arising from polarization phenomena and material transport issues.

Polarization is the deviation of the cell potential from its equilibrium value during the passage of current, which directly reduces the Voltage Efficiency. The document identifies three primary types of polarization: (1) Ohmic Polarization: Caused by the internal resistance of the battery components, including the electrolyte, the membrane, and the contact resistance between the bipolar plates and electrodes. (2) Electrochemical Polarization: Arises from the energy barrier of the redox reactions at the electrode surface. If the reaction kinetics are slow, a higher overpotential is required to drive the current. (3) Concentration Polarization: Occurs when the rate of mass transport of active species to the electrode surface is slower than the rate of reaction. This is particularly prevalent at high current densities or in "dead zones" within the flow field.

A significant challenge in RFB design is the unwanted transport of active species across the membrane, known as crossover. In non-symmetrical systems, this leads to permanent cross-contamination and capacity decay. Even in "symmetrical" systems like all-vanadium batteries, where both sides use vanadium, crossover causes self-discharge and thermal imbalances [15]. This phenomenon is heavily influenced by the membrane's selectivity and the concentration gradient between the positive and negative electrolytes. Mitigating this crossover while maintaining low Ohmic resistance remains a central focus of membrane material research.

4. Research Status and Classification of Core Membrane Materials

4.1 Functions and Key Indicators of the Separator*

The membrane, or separator, is arguably the most critical and cost-intensive component within a redox flow battery system. It serves as the physical and chemical gatekeeper of the electrochemical cell, directly influencing both the efficiency and the operational lifespan of the battery.

The primary function of the membrane is to provide a robust physical barrier that prevents the mixing of positive and negative electrolytes, which would otherwise lead to rapid self-discharge and cross-contamination. Simultaneously, the membrane must act as a selective conductor, allowing the passage of charge-carrying ions (such as H^+ or Cl^-)

to complete the internal circuit and maintain electroneutrality. This dual requirement, high ion selectivity versus low mass transport of active species, is the central challenge in membrane engineering. In aqueous systems, this often involves a complex trade-off between the hydrophilicity required for ion transport and the structural integrity needed to block bulky hydrated metal ions or organic molecules.

As illustrated in the conceptual mechanism of membrane transport (Figure 1), dense ion exchange membranes rely on fixed charged groups to "hop" carriers across the polymer matrix, while porous membranes utilize a size-sieving effect. Achieving a balance where the "gate" is wide enough for protons but too narrow for large active ions is the pinnacle of current material research.

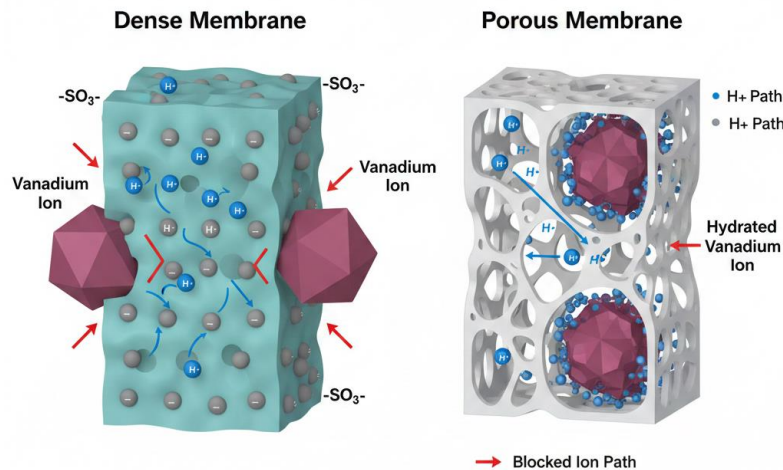


Figure 1. Illustration of ion transport mechanisms via charge-hopping in dense membranes and size-sieving in porous membranes.

To quantify the performance of a membrane, several standardized indicators are employed. The Ion Exchange Capacity (IEC) measures the concentration of fixed ionic groups within the membrane polymer, which dictates the membrane's ability to facilitate ion transport and its affinity for water. Closely related is Area Resistance, which is the inverse of ionic conductivity relative to the membrane's thickness; a lower area resistance is essential for high VE. Furthermore, the Swelling Ratio is a critical mechanical metric. It describes the volume change of the membrane upon hydration. While some swelling is necessary for ion mobility, excessive swelling can lead to dimensional instability, mechanical failure, or increased permeability to active species (crossover).

4.2 Technical Pathways for Membrane Materials*

The development of membrane materials involves several technical routes aimed at balancing cost, chemical stability, and selectivity. Each pathway employs different mechanisms to facilitate ion transport while blocking active redox species.

Dense membranes operate without macroscopic pores, relying on ion movement through polar groups within the polymer matrix. The industry standard remains Perfluorinated Sulfonic Acid (PFSA) membranes, such as Nafion, valued for their exceptional chemical stability and proton conductivity. However, PFSA membranes are limited by high costs and significant vanadium ion crossover. Consequently, non-fluorinated aromatic polymer membranes are emerging as cost-effective, eco-friendly alternatives, though they often require structural modifications like cross-linking to match the durability of PFSA materials.

In contrast, porous membranes achieve selectivity through physical size exclusion. These membranes feature interconnected networks of micropores (<2 nm), mesopores (2-50 nm), or macropores (>50 nm). By tuning the pore size to allow charge carriers through while blocking larger active species, a size-sieving effect, researchers can achieve high selectivity without expensive ion-exchange groups. Common fabrication techniques

include phase inversion and electrospinning, though maintaining a narrow pore size distribution is essential to prevent long-term active substance leakage.

A more recent innovation involves Polymers of Intrinsic Microporosity (PIMs). Unlike traditional porous membranes, PIMs utilize rigid, contorted polymer backbones that prevent efficient packing, creating "built-in" free volume or micropores (0.1--2 nm). This rigid stacking facilitates rapid ion transport while effectively blocking larger ions. While PIMs offer a high-performance, non-fluorinated solution, they currently face hurdles regarding large-scale processing and long-term stability in corrosive electrolyte environments.

5. Applications of Various Flow Battery Systems and Industrial Landscape

5.1 Prevailing Technological Pathways*

The diversification of RFB chemistry is driven by the need to balance cost, energy density, and chemical stability across different storage scales. While several chemistries have been proposed since the inception of the technology, they differ significantly in their reaction mechanisms and maturity.

The All-Vanadium Redox Flow Battery is currently the most commercially mature technology in the flow battery market. Its primary advantage lies in the use of the same element, vanadium, in different oxidation states for both the positive and negative electrolytes. This symmetrical approach effectively eliminates the problem of permanent cross-contamination; any ions that migrate across the membrane can be remediated through simple rebalancing of the electrolytes. VRFBs typically operate with a voltage window of approximately 1.25V and are capable of exceeding 15,000 to 20,000 cycles. However, the system is constrained by the relatively high cost of vanadium pentoxide and a single-electron reaction that limits its theoretical energy density.

Beyond vanadium, several alternative systems are being developed to target lower capital costs. The Iron-Chromium (Fe-Cr) system, one of the earliest flow battery technologies, utilizes abundant and inexpensive raw materials, though it faces challenges with slow reaction kinetics and hydrogen evolution. Zinc-based systems, such as Zinc-Bromine batteries, offer higher energy density due to the high solubility of zinc salts but are complicated by "phase change" issues, specifically the formation of zinc dendrites during plating which can puncture the membrane. More recently, organic flow batteries utilizing quinones or nitrogen-heterocyclic compounds have gained traction. These systems are attractive because their molecular structures can be "tuned" to optimize redox potentials and solubility, potentially allowing for multi-electron transfers that significantly boost energy density.

5.2 Industrial Status and Scalability Challenges*

The transition of RFBs from laboratory prototypes to utility-scale assets is currently underway, supported by national standards and large-scale demonstration projects. This maturation process is essential for establishing the reliability required by grid operators.

China has taken a global lead in the deployment of large-scale flow battery systems. A landmark example is the 200MW/800MWh Dalian VFB station, which completed its first phase in 2022. This project serves as a critical proof-of-concept for peak shaving and grid stabilization. Internationally, companies such as Sumitomo Electric in Japan and ESS in the United States are advancing their own proprietary systems, focusing on long-duration storage (typically 4 hours or more). These projects highlight the inherent safety of aqueous systems, which are increasingly favored for urban and industrial energy hubs where lithium-ion fire risks are a primary concern.

The industrialization of RFBs is further supported by the establishment of rigorous national and international standards. In China, standards such as GB/T 32509-2016 provide the framework for testing electrolyte purity, stack power density, and cycle stability across temperature ranges from -30C to 50C. Despite this progress, the high capital cost of the stack, specifically the perfluorinated sulfonic acid membranes, remains a barrier. Industry efforts are now focused on the localization of membrane production

and the development of non-fluorinated alternatives. As domestic production of these membranes scales, costs have already begun to drop, with some domestic perfluorinated membranes reaching 700-800 RMB per square meter. The comparison of different storage technologies provided in Table 3 contextualizes the strategic position of RFBs within the broader energy ecosystem.

Table 3. Comparison of Mainstream Energy Storage Technologies

Technology	Energy Density	Cycle Life	Primary Advantage	Typical Application
Lithium-ion	High	2,000 - 5,000	High efficiency, compact	EV, Short-term grid freq.
Pumped Hydro	Very Low	>40,000	Lowest cost per kWh	Large-scale, geography-dependent
Flow Battery	Low-Medium	15,000 - 20,000	Decoupled power/energy, safe	Long-duration grid storage
Solid-State	Very High	Ongoing R&D	High safety, high density	Future EVs, portable tech

The primary challenge remains the lower power density of flow battery stacks compared to lithium-ion counterparts, which results in a larger physical footprint. To address this, current research is moving toward multi-electron active species and high-concentration electrolytes to improve volumetric energy density. Furthermore, the development of standardized "plug-and-play" containerized modules is expected to simplify the deployment of hundred-megawatt stations. As the market moves toward longer discharge durations (6-12 hours), the low marginal cost of adding electrolyte tanks ensures that RFBs will become increasingly competitive against lithium-ion systems for seasonal and long-term storage needs.

6. Conclusion and Outlook

The transition toward a sustainable energy landscape necessitates storage solutions that prioritize safety, longevity, and scalability. As evidenced throughout this study, redox flow batteries represent a transformative technology specifically engineered for the demands of large-scale, long-duration energy storage. By decoupling power and capacity through an external electrolyte circulation system, flow batteries overcome the inherent physical constraints of traditional closed-cell architectures like lithium-ion systems. The inherent safety of aqueous media, which eliminates the risk of thermal runaway and catastrophic combustion, positions flow batteries as the most viable candidate for massive grid-level installations. Furthermore, the exceptional cycle life of up to 20,000 cycles ensures a lower leveled cost of storage over the system's operational lifetime, despite the current higher initial capital expenditure.

However, the widespread commercialization of flow batteries still faces several critical technical and economic hurdles. The reliance on expensive perfluorinated sulfonic acid membranes and the limited energy density of single-electron metal ion electrolytes remain the primary bottlenecks. Moving forward, the industry must focus on three strategic pillars: material innovation, cost reduction, and system standardization. The development of non-fluorinated, low-cost ion exchange membranes and precisely engineered porous separators will be essential to reduce stack costs and environmental impact. Simultaneously, exploring multi-electron organic molecules and high-solubility active substances offers a promising pathway to enhance volumetric energy density, potentially expanding the application of flow batteries beyond stationary storage.

In conclusion, while lithium-ion batteries will continue to dominate short-term and portable power markets, redox flow batteries are poised to become the backbone of long-term grid stability. The successful deployment of hundred-megawatt-level demonstration projects and the ongoing establishment of national standards underscore the maturity of this technology. By addressing the current challenges in membrane selectivity and electrolyte concentration through interdisciplinary research, the energy sector can realize a resilient and flexible power grid. The future of energy storage lies not in a single dominant technology, but in a diversified ecosystem where the unique safety and scalability of flow batteries play an indispensable role in achieving global carbon neutrality goals.

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