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Increasing energy density of vanadium redox flow batteries: A comprehensive review

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ABSTRACT

Background: Vanadium Redox Flow Batteries (VRFBs) represent a leading energy storage technology for renewable integration due to their long cycle life, high safety, and flexible scalability. However, their low energy density and high cost continue to limit widespread adoption. This study aims to synthesize and critically evaluate recent advances in enhancing VRFB performance through innovations in electrode materials, electrolyte chemistry, and membrane design. **Methods:** This study adopts a comprehensive literature review approach, analyzing theoretical and experimental research published in recent years. The review focuses on advancements in nanostructured electrode surfaces, optimized electrolyte formulations, and functional hybrid membranes. Theoretical insights from materials science and electrochemistry were integrated to establish the correlation between structure, performance, and efficiency. **Findings:** The reviewed studies reveal that nanostructured and heteroatom-doped electrodes enhance redox kinetics and minimize side reactions, while optimized electrolytes with mixed acids and stabilizers improve vanadium solubility and thermal stability. Hybrid polymer–inorganic membranes effectively reduce vanadium ion crossover and maintain high proton conductivity, thereby increasing coulombic and energy efficiencies. Collectively, these advancements improve power output, reduce self-discharge, and enhance long-term cycling performance, moving VRFBs closer to economic feasibility. **Conclusion:** Advancements in material design and system optimization are pivotal in overcoming the limitations of conventional VRFBs. Continued research on scalable, low-cost materials, electrolyte recycling, and hybrid integration will further promote sustainable energy storage. **Novelty/Originality of this article:** This review uniquely integrates material-level and system-level perspectives, offering a holistic understanding of how innovations across components collectively advance high-efficiency, cost-effective, and environmentally sustainable VRFB technology for next-generation renewable energy systems.

KEYWORDS: vanadium redox flow battery (VRFB); energy density; electrolyte optimization; electrode modification; membrane engineering.

1. Introduction

Climate change has emerged as one of the most important environmental concerns of our time, with significant correlations between greenhouse gas emissions and their negative impact on nature (Hu et al., 2022). As energy consumption climbs and fossil fuel reserves degrade, renewable energy has emerged as a vital answer for long-term energy supply. With global efforts to reduce carbon emissions and mitigate climate change, renewable energy sources, have received significant attention for their low environmental impact and potential to meet rising energy demands while reducing the carbon footprint of electricity generation (Skyllas-Kazacos et al., 2011).

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The global shift toward renewable energy is evident from production trends: since 2011, total electricity generation has increased by an average of 2.4% annually, while renewable sources have expanded much faster, at about 6.1% per year compared to only 1.3% for non-renewables. In 2022 alone, renewable electricity generation rose by 7.2% relative to 2021, driven by solar and wind power, which together made up 11.7% of global electricity, an 18.2% increase from the previous year (IRENA, 2024).

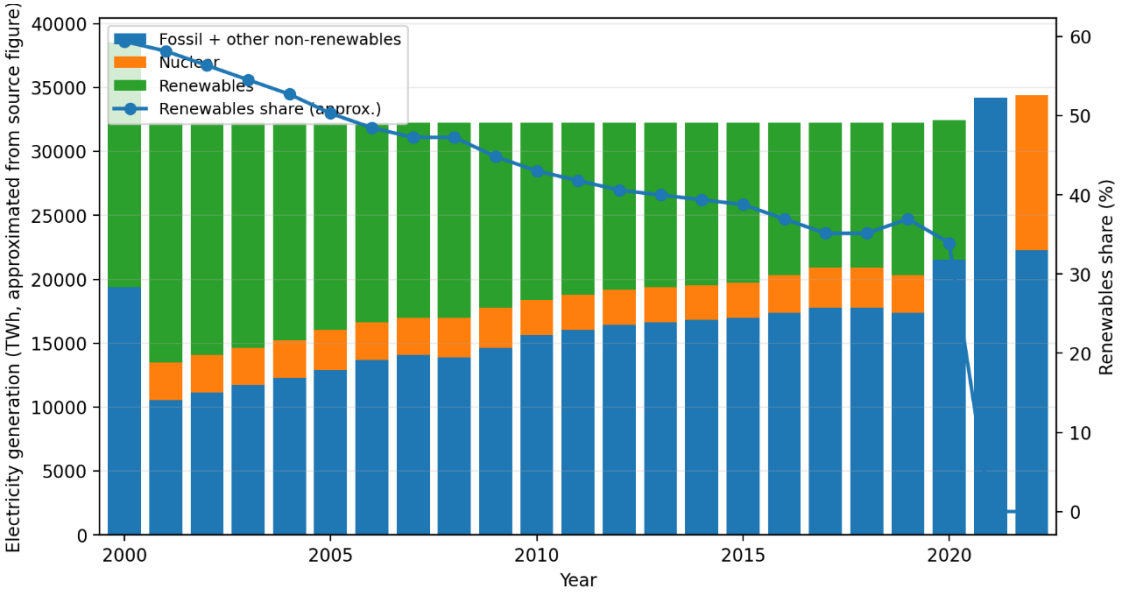

Fig. 1. Comparison of average annual growth rates in global electricity generation since 2011, redrawn and adapted from data reported by the International Renewable Energy Agency
(IRENA, 2024)

Renewable energy sources such as solar and wind are inherently variable and unpredictable, leading to fluctuations in power generation. Their non-dispatchable nature makes it challenging to ensure a stable electricity supply, as they cannot be easily adjusted to match real-time demand (Skyllas-Kazacos et al., 1991). To overcome this issue, energy storage systems (ESS) play a key role in grid stabilization by holding extra energy during periods of high generation and releasing it when required. Although batteries have long been recognized for their importance in remote area power supply (RAPS) and renewable energy storage (Stark et al., 2015). Their high cost has limited their broad application. However, with the increasing usage of renewable energy and the growing requirement for a consistent power supply, the focus has now shifted toward developing energy-efficient, safe, and cost-effective storage solutions (Cotterman, 2013).

As the transition to renewable energy accelerates, the intermittent nature of these sources creates a growing need for energy storage systems capable of balancing supply and demand in real time. Although pumped hydro energy storage (PHES) has traditionally been the leading technology for large-scale storage (Chen et al., 2013c). Its drawbacks, such as extensive land requirements, high capital costs, and environmental concerns, limit its suitability for managing the increasing variability of renewable power generation (Wang et al., 2013b). On the other hand, Redox flow batteries (RFBs) have garnered growing interest for energy storage system (ESS) applications. A key distinguishing feature of RFBs is the spatial separation between energy storage and energy conversion, a characteristic not achievable in other secondary batteries that rely on solid-state active materials (Li et al., 2024a).

Redox Flow Batteries (RFBs) have gained attention as a promising technology for large-scale energy storage, offering several advantages that make them well-suited for grid applications (Leung et al., 2012). They operate using two separate electrolyte solutions containing distinct redox couples, which are circulated through an electrochemical cell

stack to enable energy storage and conversion (Skyllas-Kazacos et al., 2010). The modular configuration of RFBs provides operational flexibility, as energy capacity depends on the electrolyte volume, while the power output is determined by the number of cells in the stack (Ye et al., 2017). RFBs are perfectly scalable due to their independence from energy and power capacity, allowing them to efficiently meet a wide range of energy demands (Houser et al., 2017). Furthermore, RFBs have a long operational life and minimal maintenance requirements due to their basic design, which comprises a few moving components and quick response times. Their capacity to fully charge and discharge without significant degradation of cell components offers them a theoretically limitless life cycle, which adds to their attraction for renewable energy storage (De Leon et al., 2006). Furthermore, compared to other new energy storage technologies, RFBs have reduced operational and capital costs, especially in large-scale applications (Skyllas-Kazacos et al., 2011).

Among the different redox flow battery chemistries, the Iron–Chromium Redox Flow Battery (ICRFB) represents a well-established aqueous system that employs Fe^{2+}/Fe^{3+} and Cr^{3+}/Cr^{2+} redox couples in a hydrochloric acid electrolyte. It provides a standard cell voltage of approximately 1.18 V and offers advantages such as the abundance and low cost of its active materials (Zhang et al., 2017). Innovations such as interdigitated flow fields (IFF) have been shown to enhance mass transport and improve energy efficiency to over 80% at high current densities (Zeng et al., 2016). Additionally, electrocatalysts like bismuth-doped carbon (Bi-C) have helped address the slow kinetics of the Cr^{3+}/Cr^{2+} couple and reduce hydrogen evolution (Ahn et al., 2021). Electrochemical purification methods help minimize impurity-induced side reactions, thereby enhancing cycle stability (Wan et al., 2023). Nevertheless, ICRFBs continue to face several challenges, including hydrogen evolution, catalyst degradation, ion crossover through the membrane, and the need for high operating temperatures, factors that collectively hinder their large-scale implementation.

Another notable system is the Zinc–Bromine Redox Flow Battery (ZBFB), a hybrid configuration in which zinc is electrodeposited during charging and bromine is generated at the positive electrode. With a higher cell voltage (~1.8 V) and energy density reaching up to 70 Wh/kg, ZBFBs are known for their cost-effectiveness and the accessibility of zinc and bromine (Jiménez-Blasco et al., 2023). Their architecture enables partial decoupling of power and capacity, making them attractive for stationary storage. Compared to vanadium RFBs, ZBFBs benefit from significantly lower electrolyte costs and higher cell voltage. Moreover, improvements in membrane design, such as the use of amphoteric functionalized silica (Am-SiO_2) in Nafion, have increased coulombic efficiency to 99.4% by suppressing bromine crossover while enabling bi-ionic transport of Zn^{2+} and Br^-(Han & Shanmugam, 2022). Carbon-based positive electrodes have also been optimized for corrosion resistance and redox kinetics(Popat et al., 2022). However, ZBFBs face several operational challenges, including bromine toxicity and volatility, which necessitate the use of complexing agents to prevent evaporation and crossover. Additionally, the formation of dense polybromide phases and poor electrolyte mixing can lead to zinc dendrite formation, ultimately compromising cycle stability and safety (Han & Shanmugam, 2022).

In addition to these, other redox chemistries such as Zinc–Cerium, All-Iron, and Hydrogen–Bromine RFBs are under active exploration for their unique advantages. Zinc–Cerium RFBs achieve some of the highest voltages among aqueous systems (up to 2.4 V) by pairing Zn/Zn^{2+} and Ce^{3+}/Ce^{4+} couples, enabling high power output. However, sluggish zinc kinetics and considerable voltage losses from ohmic and mass transfer resistances have limited their performance, often requires operation at elevated temperatures (Amini & Pritzker, 2020). All-Iron RFBs, which rely on Fe^{2+}/Fe^0 and Fe^{3+}/Fe^{2+} couples, stand out for their low-cost, sulfate-based electrolytes and compatibility with microporous membranes. The addition of EMIC has improved iron solubility and plating behavior, extending cycle life above 800 cycles. Yet, real-time operation is complicated by pH imbalance between half-cells, hydrogen evolution, and membrane fouling (Yu et al., 2021). Meanwhile, Hydrogen–Bromine RFBs offer extremely high energy densities (~195.8 Wh/L) and fast kinetics, using HBr/Br_2 electrolytes and hydrogen gas electrodes. Although bromine complexing agents like $[C_2Py]Br$ help suppress volatility, they also reduce conductivity and introduce phase

separation and membrane degradation, limiting usable capacity to just 30% during prolonged cycling (Küttinger et al., 2021).

While various redox flow battery chemistries offer unique advantages and face distinct technical challenges, one system has consistently stood out in both research and commercialization: the Vanadium Redox Flow Battery (VRFB). Its widespread appeal stems from the use of a single electroactive element, vanadium, in multiple oxidation states, which reduces the risk of cross-contamination and enhances long-term cycling stability. Unlike conventional batteries that store energy in solid electrodes, VRFBs store energy in liquid electrolytes, housed in external tanks and circulated through an electrochemical cell. This architecture not only allows independent scaling of energy and power but also supports high operational flexibility, deep discharge capability, and extended service life (Skyllas-Kazacos et al., 2010). As a result, VRFBs have become a leading candidate for grid-level energy storage, particularly in applications requiring durability, scalability, and safety.

2. Methods

This study adopts a comprehensive literature review approach to evaluate recent strategies for increasing the energy density of vanadium redox flow batteries (VRFBs). Relevant articles were collected from major scientific databases, focusing on recent peer-reviewed studies that report advancements in electrode materials, electrolyte systems, and membrane technologies.

The selected literature was categorized into three main areas: electrode modification, electrolyte optimization, and membrane development. Comparative analysis was conducted to assess the impact of these approaches on key performance parameters, including energy efficiency and cycling stability. In addition, fundamental concepts from electrochemistry and materials science were used to interpret the relationship between material properties and battery performance. Only studies with clearly reported methodologies and reliable data were included to ensure the validity of the analysis.

3. Results and Discussion

3.1 Vanadium redox flow batteries

Vanadium Redox Flow Batteries (VRFBs) are among the most extensively studied and implemented redox flow battery systems, recognized for their scalability, long cycle life, and effectiveness in grid-scale energy storage applications (Wang et al., 2013b). Unlike conventional solid-state batteries, VRFBs store energy in externally circulated liquid electrolytes (Bartolozzi, 1989). These electrolytes contain vanadium ions in multiple oxidation states, allowing energy storage and release through reversible redox reactions (Guarnieri et al., 2016). A major advantage of VRFBs lies in the decoupling of power and energy: the power output is determined by the size and number of cell stacks, whereas the energy capacity depends on the volume of electrolyte in the storage tanks. This modular flexibility makes VRFBs suitable for both short- and long-duration energy storage applications (Rahman & Skyllas-Kazacos, 2009).

What sets VRFBs apart from other flow battery chemistries is their use of the same element, vanadium, in both half-cells, but in four distinct oxidation states (V^{2+}, V^{3+}, VO^{2+}, and VO_2^{+}) (Gundlapalli et al., 2018). This design choice significantly reduces the risk of cross-contamination between electrolytes, a frequent problem in other systems, thereby improving long-term stability and simplifying maintenance (Skyllas-Kazacos & Kazacos, 2011). In the event of minor crossover, performance degradation is minimal and can often be reversed by simply remixing the electrolytes. Moreover, since vanadium-based systems avoid the need for different redox pairs, they eliminate issues related to mismatched solubilities or membrane selectivity, which are often limiting factors in other RFB chemistries. VRFBs also benefit from excellent operational durability (Sum et al., 1985). Because they undergo minimal structural degradation during cycling, their service life often

exceeds 10,000 cycles with stable capacity retention. Furthermore, they are considered inherently safe systems due to their non-flammable aqueous electrolytes and low-pressure operation (Soloveichik, 2015). As a result, VRFBs have gained significant attention for applications in renewable energy integration, load levelling, and backup power, especially where reliability, safety, and long-term economics are critical.

3.1.1 Working Principle

The operation of a Vanadium Redox Flow Battery (VRFB) relies on the controlled transfer of vanadium ions in different oxidation states between two distinct electrolyte circuits, driven by external pumps and a central electrochemical stack (de León et al., 2006). During operation, electrolytes from the positive and negative storage tanks are continuously circulated through the stack, where redox reactions occur at porous carbon-based electrodes (Arevalo-Cid et al., 2021).

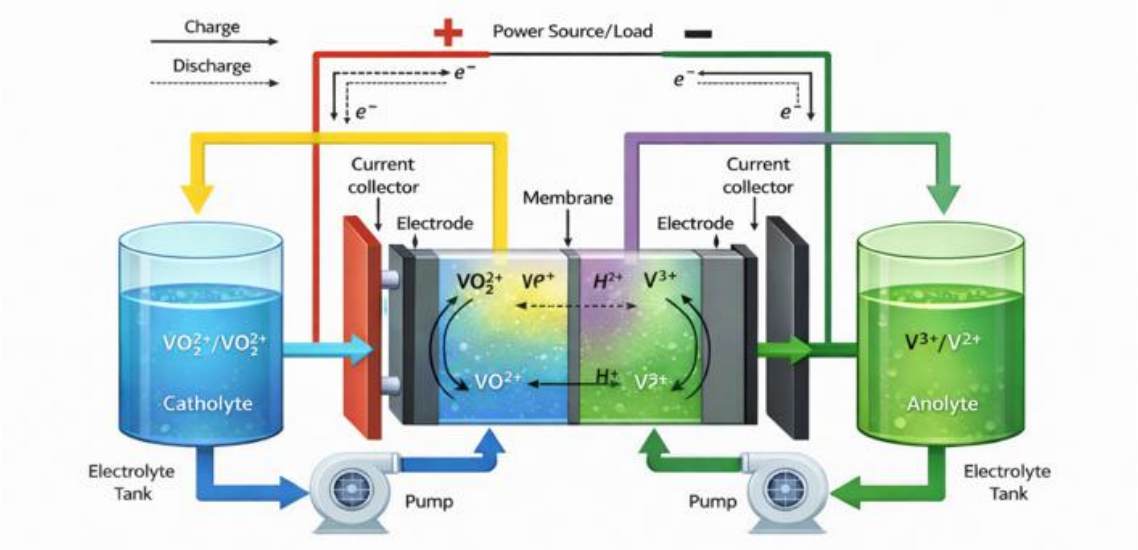

Fig. 2. Schematic illustration of the working principle of a vanadium redox flow battery (VRFB), adapted from (Puleston et al., 2022) under CC BY 4.0

Within the stack, each half-cell is equipped with an electrode and separated by a proton exchange membrane (PEM), which permits the passage of protons (H^+) to maintain charge balance while preventing physical mixing of the two electrolytes. During charging, an external power source induces oxidation and reduction reactions: V^{3+} ions in the negative electrolyte are reduced to V^{2+}, whereas VO^{2+} ions in the positive electrolyte are oxidized to VO_2^+. These half-cell reactions are represented as:

Negative electrode (reduction):

$$V^{3+} + e^- \rightarrow V^{2+} \quad \text{(Eq. 1)}$$

Positive electrode (oxidation):

$$VO_2^+ + 2H^+ - e^- \rightarrow VO^{2+} + H_2O \quad \text{(Eq. 2)}$$

Conversely, during discharge, the redox reactions proceed in the opposite direction. V^{2+} is oxidized back to V^{3+} at the negative electrode, and VO_2^+ is reduced to VO^{2+} at the positive electrode, releasing electrical energy to the external circuit (Gundlapalli et al., 2018). The system is highly modular, and several individual cells are connected in series within the stack using bipolar plates (Arenas et al., 2017). These plates help in current collection and

mechanical support, enabling uniform distribution of electrolyte and maintaining consistent flow. Since the redox reactions take place only at the electrode surfaces, and the electrodes themselves are chemically inert, the battery demonstrates excellent reversibility and long-term operational stability. The amount of energy stored is determined by the volume and concentration of active vanadium species in the electrolyte, while voltage is defined by the electrochemical potential between the two half-cells(Jirabovornwisut & Arpornwichanop, 2019). Typically, the cell delivers an open-circuit voltage of around 1.25 V. To achieve higher system voltages and capacities, multiple stacks can be combined depending on the specific application (Skyllas-Kazacos & Grossmith, 1987).

3.1.2 Recent advancement in Vanadium redox flow batteries

Early generations of Vanadium Redox Flow Batteries (VRFBs), though promising for long-duration energy storage, were hindered by critical limitations such as low energy density, poor reaction kinetics at the electrodes, membrane degradation under prolonged cycling, and high system costs (Leung et al., 2012; Trovò et al., 2019). The sluggish electrochemical activity of vanadium redox couples, especially at carbon-based electrodes, resulted in considerable activation losses and limited the overall power output (Davies & Tummino, 2018). In addition, conventional porous fiber felt electrodes (PFFEs), while widely used, suffered from low catalytic activity and insufficient active sites, which further reduced the system's energy efficiency and response speed. The supporting membrane materials were prone to chemical attack from V^{5+} ions, causing reduced selectivity and ion crossover over time, which compromised coulombic efficiency and cycle life (Ding et al., 2023).

To address these challenges, recent advancements have targeted both macroscopic and microscopic redesign of key VRFB components. On the electrode side, next-generation PFFEs have been improved through multiscale structural optimization, including compression tuning, hierarchical porosity engineering, and shape reshaping via computational flow field modelling (Aramendia et al., 2021). These changes enhance mass transport, reduce polarization, and enable more uniform electrolyte distribution across the electrode surface. At the micro level, surface modification with oxygen-containing functional groups and the introduction of nanocatalysts, such as graphene oxide, carbon nanotubes, and high-entropy oxides, have significantly accelerated redox kinetics by increasing the number of active sites and reducing charge transfer resistance (Li et al., 2024b).

Moreover, research has improved electrolyte stability through optimized vanadium concentrations and temperature control strategies to prevent precipitation, allowing higher energy densities up to 35 Wh/kg under stable cycling conditions. The system architecture has also evolved with modular cell-stack designs that support higher current densities (up to 400 mA cm^{-2}), thereby reducing stack size and improving power density, lowering system costs by up to 30–40%. Collectively, these advancements mark a transformative shift from early, lab-scale VRFB prototypes toward commercially viable systems with enhanced efficiency, scalability, and economic competitiveness for grid-scale deployment (Viswanathan et al., 2023).

3.2 Modification in VRFBS

Despite advancements in VRFB technology, improving energy density remains an important topic of study. Several variables impact the decrease in energy density of vanadium redox flow batteries (VRFBs), including the electrode, electrolyte, and membrane that separates the electrolyte. The restricted solubility of vanadium ions in H_2SO_4 + H_2O solution limits the quantity of vanadium accessible for redox reactions, limiting energy density (Vijayakumar et al., 2015). At high temperatures, vanadium species, especially V^{5+}, become unstable and produce irreversible V_2O_5 precipitates, reducing energy storage capability (Kausar et al., 2016).

Finally, the membrane that separates the electrolytes is crucial for preventing cross-contamination between positive and negative electrolytes. However, inefficiencies in ion selectivity or greater resistance across the membrane might cause greater energy losses during operation, thus reducing total energy density (Chen, 2023). These factors, electrolyte solubility and stability, electrode performance, and membrane efficiency, all contribute to the issue of increasing energy density in VRFBs.

3.2.1 Electrode

In vanadium redox flow batteries (VRFBs), the electrode is critical to system performance, including power density, energy density, and energy efficiency. The electrode is a place for redox reactions, providing an electroactive surface and aiding electron conduction required for electrochemical processes(Park et al., 2014). Carbonaceous materials, such as carbon and graphite felt, are the most often employed electrodes in vanadium redox flow batteries (VRFBs) because to their high electrical conductivity, chemical stability, and corrosion resistance (Kim et al., 2015). To enhance the energy density of VRFBS, the following methods can be used.

3.2.1.1 Anode

Yang and colleagues demonstrated that modifying graphite felt electrodes with Nb_2CT_x MXene significantly improved the kinetics of the V^{3+}/V^{2+} redox reaction, a key limitation in VRFB anode performance. Their experimental results showed that the Nb_2CT_x-based electrode achieved an energy efficiency of 83.1% at 160 mA cm^{-2}, 12.3% higher than that of pristine graphite felt. The enhanced performance was attributed to the high catalytic activity and hydrogen evolution reaction (HER) suppression capabilities of Nb_2CT_x, as confirmed through density functional theory (DFT) calculations. These findings position Nb-based MXenes as promising electrocatalysts for improving energy density and cycling stability in VRFB systems (Tiwari et al., 2024).

The research conducted by Wu et al. (2014) introduces a high entropy alloy oxide (HEAO) coating strategy on graphite felt anodes using high power impulse magnetron sputtering (HiPIMS), aiming to enhance the performance of vanadium redox flow batteries (VRFBs). The uniform deposition of amorphous VNbMoTaWOx on the graphite felt significantly boosts the VO_2^+/VO^{2+} redox reaction kinetics due to the presence of abundant oxygen vacancies and multi-valence metal ions. This anode modification leads to a remarkable increase in energy efficiency, reaching 81.40% at 100 mA cm^{-2} and 71.40% at 160 mA cm^{-2}, an improvement of 7.70% and 17.30% respectively over the uncoated electrode. The enhanced electrochemical activity and ion transport are attributed to the synergistic effects of uniform coating thickness, surface conductivity, and the creation of high-density active sites, making this approach a promising pathway for advancing large-scale VRFB systems (Li et al., 2025).

A dual-doping strategy using nitrogen and oxygen to construct a porous electrode by electrochemically depositing ZIF-8 derivatives on pristine graphite felt. This innovative approach significantly improved the redox kinetics of both the V^{2+}/V^{3+} and VO_2^+/VO^{2+} couples, addressing the sluggish kinetics that traditionally limit the VRFB performance. The doped porous structure enhanced the reactive surface area and electron transfer, resulting in a substantial boost in battery performance. The modified VRFB achieved a high energy efficiency of 85.77% at 100 mA cm^{-2} and showed stable cycling for 500 cycles, along with a peak power density of 356 mW cm^{-2}(Lobato-Peralta et al., 2025).

Furthermore, an innovative approach using high-power impulse magnetron sputtering (HiPIMS) to deposit a uniform layer of VNbMoTaW high-entropy alloy oxide (HEAO) onto graphite felt electrodes. This modification significantly enhanced the electrode's surface hydrophilicity and introduced abundant oxygen vacancies, which collectively accelerated the kinetics of vanadium redox reactions. The amorphous structure of the HEAO coating further improved mass transport by reducing ion diffusion distances. As a result, the

modified electrode achieved a notable energy efficiency of 80.50% at 100 mA cm^{-2}, representing a 9.49% improvement over the pristine electrode, highlighting the role of multimetal oxide coatings in optimizing both anode activity and energy density in VRFBs (Ren et al., 2023).

Wang et al. introduced a dual-doped strategy to enhance anode performance in vanadium redox flow batteries (VRFBs) by co-doping nitrogen and oxygen into porous carbon frameworks. The doped porous electrodes were synthesized by electrochemically depositing ZIF-8 derivatives on graphite felt, resulting in a high surface area with abundant active sites for vanadium redox reactions. This modification significantly improved the electrochemical kinetics of both V^{3+}/V^{2+} and $VO_2{}^+/VO^{2+}$ couples. The enhanced redox activity enabled the modified VRFB to achieve an impressive energy efficiency of 85.77% at 100 mA cm^{-2} and a stable cycling performance over 500 cycles. Additionally, a peak power density of 356 mW cm^{-2} was observed, underscoring the positive impact of the dual-doped structure on energy density (Jiang et al., 2024).

In a recent study, Yao et al. introduced an innovative in-situ electrodeposition strategy to enhance the anode performance of vanadium redox flow batteries. By using a low-concentration V^{3+} catholyte (33 mM) during the electrodeposition process, they successfully deposited uniform and dense bismuth nanoparticles (~58 nm) onto the graphite felt anode. This approach prevented oxidation of Bi nanoparticles by avoiding high $VO_2{}^+$ crossover, which is common in conventional methods. The resulting modified anode demonstrated superior catalytic activity toward the V^{3+}/V^{2+} redox couple, leading to an energy efficiency of 76.3% at 300 mA cm^{-2}, outperforming both conventionally treated and untreated electrodes. Additionally, the battery maintained a high energy efficiency of 82.5% over 200 cycles at 200 mA cm^{-2}, highlighting the potential of Bi-modified anodes to improve both reaction kinetics and long-term performance in VRFBs (Chen et al., 2025).

Zhang et al. developed a novel anode material for vanadium redox flow batteries by electrodepositing Bi nanoparticles uniformly onto graphite fibers using a low-concentration vanadium ion catholyte. This in-situ deposition strategy significantly minimized the oxidative degradation typically caused by $VO_2{}^+$ crossover from the catholyte, resulting in Bi nanoparticles of approximately 58 nm evenly distributed across a large-scale anode area. The enhanced electrode demonstrated a notable energy efficiency of 76.3% at 300 mA cm^{-2}, surpassing both conventionally treated (74.9%) and untreated electrodes (73.3%), and achieved 82.5% energy efficiency over 200 stable cycles at 200 mA cm^{-2} (Mizrak et al., 2025).

Duan et al. proposed an effective anode modification strategy by introducing bismuth nanoparticles onto graphite felt electrodes to enhance the sluggish kinetics of the V^{2+}/V^{3+} redox couple in vanadium redox flow batteries. The uniform and dense deposition of Bi nanoparticles significantly improved charge-transfer characteristics, leading to enhanced electrochemical activity and reduced overpotential at the anode. The optimized Bi-modified electrode delivered higher energy efficiency and excellent cycling stability compared to pristine graphite felt, demonstrating the strong potential of metal nanoparticle decoration in overcoming anode limitations. This approach highlights a promising pathway to increase the energy density and long-term viability of VRFBs through rational anode engineering (Duan et al., 2019).

Figure 3 summarizes the mechanistic pathways through which diverse anode engineering strategies enhance the electrochemical performance of vanadium redox flow batteries. Although MXene coatings, high-entropy alloy oxide films, dual-doped porous carbon frameworks, and metal nanoparticle decorations differ in composition and fabrication routes, they exhibit a clear mechanistic convergence. These approaches collectively accelerate the charge-transfer kinetics of the V^{3+}/V^{2+} redox couple by increasing the density of accessible active sites, improving electrode wettability, and facilitating rapid electron transport at the electrode–electrolyte interface.

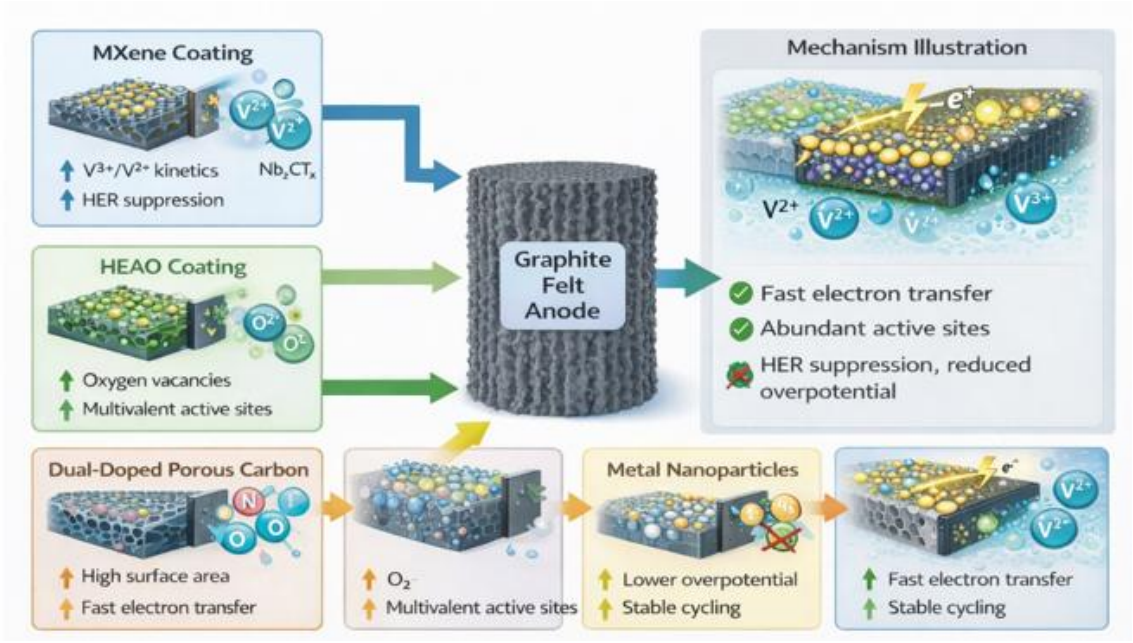

Fig. 3. Mechanistic overview of advanced anode engineering strategies for vanadium redox flow batteries

The introduction of oxygen vacancies, heteroatom dopants, and multivalent metal centers further strengthens vanadium ion adsorption while lowering charge-transfer resistance and reaction overpotential. Importantly, several surface modification strategies effectively suppress the competing hydrogen evolution reaction, particularly under high current densities, thereby enhancing energy efficiency and cycling stability. This mechanistic synergy underscores rational anode surface engineering as a critical design principle for overcoming kinetic limitations and advancing the energy density, durability, and scalability of next-generation VRFB systems.

3.2.1.2 Cathode modification

Flox et al. (2013) investigated the strategies to enhance the electrochemical activity of carbon-based electrodes in all-vanadium redox flow batteries (VRFBs). The study focused on improving the sluggish kinetics of the positive electrode (VO_2^+/VO_2^{++} redox couple) by modifying carbon materials. Two approaches were employed: (i) decorating graphene substrates with bimetallic $CuPt_3$ nanocubes, and (ii) thermally treating polyacrylonitrile (PAN) felt electrodes under NH_3/O_2 atmosphere to introduce nitrogen- and oxygen-containing functional groups. Both modifications significantly enhanced electron and oxygen transfer processes, yielding higher current densities, reduced charge-transfer resistance, and improved energy efficiency. PAN electrodes treated at 500 °C for 24 h demonstrated ~84% energy efficiency over 30 cycles at 20 mA cm^{-2}. These results highlight the effectiveness of cathode-side electrode engineering in addressing kinetic limitations and improving VRFB performance. Furthermore, González et al. developed a graphene-modified graphite felt (GF-G) using electrophoretic deposition of graphene oxide (GO) suspensions to enhance VRFB performance. The modification yielded a 3D hybrid architecture of felt fibers interconnected with reduced graphene oxide sheets decorated by abundant oxygen-containing functional groups. As a positive electrode, GF-G exhibited lower charge-transfer resistance and quasi-reversible redox kinetics for the VO_2^+/VO_2^{++} couple. In single-cell VRFB tests at 25 mA cm^{-2}, GF-G delivered a discharge capacity of 14.1 Ah L^{-1} and an exceptionally high energy efficiency of 95.8%, outperforming untreated and thermally treated graphite felt (González et al., 2017).

Xiang & Daoud (2019) reported a facile cobalt oxide (CoO) modification of graphite felt (GF) electrodes via impregnation and ultrasonication, followed by calcination, to improve

VRFB performance. The CoO-modified felt (CoTGF) exhibited a rough, ultra-thin coating layer that significantly enhanced surface area and active sites for the VO_2^+/VO_2^{++} redox couple. Electrochemical analyses showed improved reversibility (ΔEp reduced to 216 mV), reduced charge-transfer resistance, and higher diffusion capacitance. In single-cell tests at 150 mA cm^{-2}, CoTGF achieved an energy efficiency of 69.4% and a discharge capacity of 373.9 mAh, representing ~12.7% and ~101.7% improvements over pristine GF, respectively. This demonstrates that cobalt oxide modification is a simple and effective strategy to boost cathode kinetics and overall energy density in VRFBs. Grosse Austing introduced a novel unitized bidirectional vanadium/air redox flow battery (VARFB) that replaces the conventional VO_2^+/VO_2^{++} couple with the O_2/H_2O redox reaction, thereby increasing the cell potential from 1.25 V to 1.49 V and roughly doubling the theoretical energy density. A two-layered cathode design was employed: a Pt/C-based gas diffusion electrode for the oxygen reduction reaction during discharge, combined with an IrO_2-modified graphite felt for the oxygen evolution reaction during charge. In single-cell tests, the VARFB achieved a maximum energy efficiency of 41.7% at 20 mA cm^{-2} and an average discharge power density of 34.6 mW cm^{-2} at 40 mA cm^{-2} (Grosse Austing et al., 2015).

Xiang & Daoud (2018)developed a facile impregnation–ultrasonication–calcination strategy to deposit an ultra-thin Cr_2O_3 coating onto graphite felt (GF) electrodes for VRFBs. The Cr_2O_3-modified GF exhibited enhanced wettability, reduced charge-transfer resistance, and quasi-reversible kinetics for the VO_2^+/VO_2^{++} redox couple. In single-cell tests, the modified electrode achieved a voltage efficiency of 75.9% and energy efficiency of 67.6% at 150 mA cm^{-2}, representing substantial improvements over pristine GF. The discharge capacity increased by 83.6%, and the electrode maintained stable performance after 50 cycles, highlighting the durability of the coating. These results demonstrate that chromium oxide modification is an effective, low-cost strategy to improve cathode kinetics and energy efficiency in VRFBs.

Xia et al. (2019) fabricated a graphene-coated carbon felt (G/CF) electrode for VRFBs using a simple solution coating method. The graphene layer provided a high surface area and superior conductivity, significantly enhancing the electrochemical activity of the VO_2^+/VO_2^{++} redox couple. Optimal dipping (five times) yielded the best catalytic behavior with a reduced peak potential difference and improved reversibility. In single-cell VRFB tests, G/CF electrodes delivered consistently higher voltage and energy efficiencies compared to pristine CF, remaining above 80% even after 500 cycles at 80 mA cm^{-2}. The modification also increased the discharge capacity stability and raised the peak power density by 39 mW cm^{-2}. These results highlight graphene coating as a cost-effective and scalable strategy for boosting cathode kinetics and overall VRFB efficiency.

Bayeh et al. (2018) synthesized Ta_2O_5 nanoparticle-modified graphite felt (Ta_2O_5-GF) electrodes via a one-step hydrothermal method for VRFBs. Uniformly distributed hexagonal Ta_2O_5 nanoparticles significantly increased surface oxygen-containing groups and improved wettability, creating abundant active sites for the VO_2^+/VO_2^{++} redox couple. The optimized 0.75 wt% Ta_2O_5-GF electrode exhibited superior kinetics with reduced charge-transfer resistance (13.1 Ω vs. 47.3 Ω for pristine GF). In single-cell tests at 80 mA cm^{-2}, it achieved a coulombic efficiency of 94.8%, a voltage efficiency of 78%, and an energy efficiency of 73.7%, representing a ~9% EE improvement over unmodified GF. After 100 cycles, efficiencies remained stable, confirming excellent durability.

Cathode modification in vanadium redox flow batteries primarily targets the sluggish kinetics of the VO_2^+/VO^{2+} redox couple by increasing electrochemically active surface area, enhancing electron transfer, and introducing catalytically active sites. Strategies such as graphene or reduced graphene oxide coatings improve electrical conductivity and accessible reaction sites, while heteroatom doping (e.g., N, O) and metal or metal-oxide decorations (e.g., $CuPt_3$, CoO, Cr_2O_3, Ta_2O_5) accelerate interfacial charge transfer and improve electrode wettability. These modifications collectively reduce polarization losses and charge-transfer resistance, enabling higher operating current densities and improved voltage efficiency. As a result, enhanced cathode kinetics directly translate into increased

usable capacity, higher power output, and improved energy density at the cell level, thereby strengthening the viability of VRFBs for high-performance grid-scale energy storage.

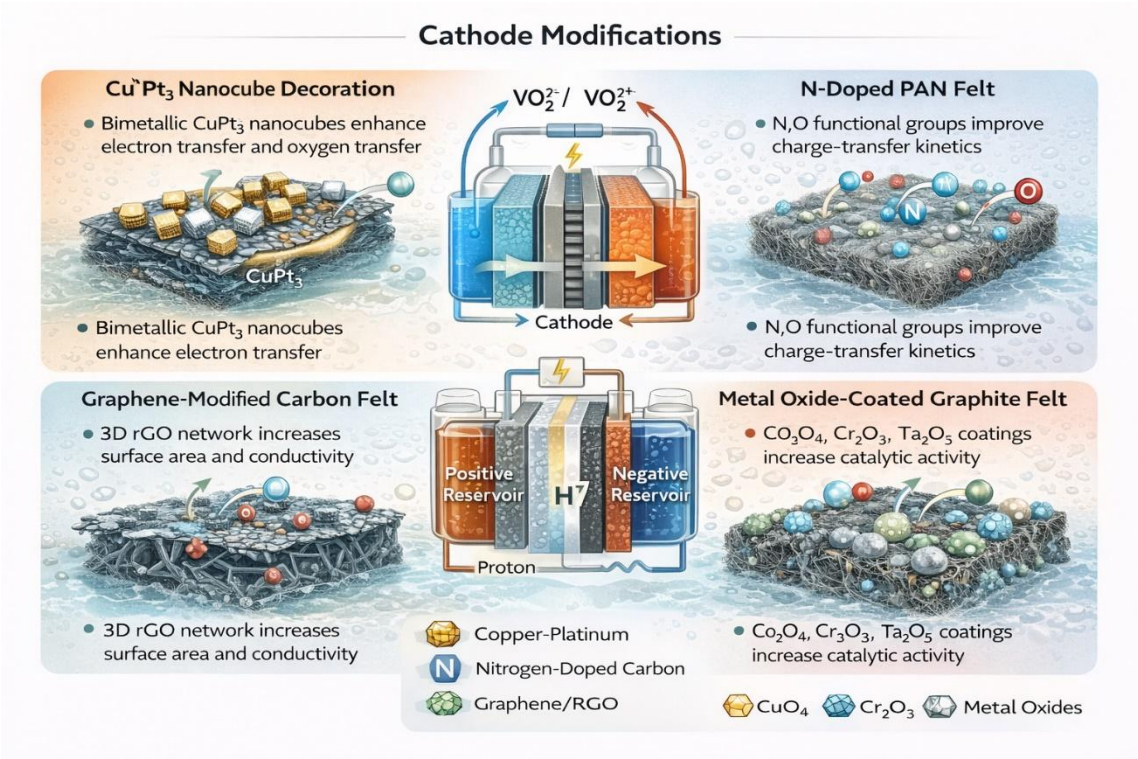

Fig. 4. Overview of cathode modification strategies for vanadium redox flow batteries

3.2.2 Electrolyte

There are several difficulties associated with the electrolytes that cause VRFBs to have a lower energy density. At elevated temperatures, vanadium ions, particularly V(V), precipitate as V_2O_5 and decrease reversibility due to their restricted solubility. Because V(II), V(III), and V(IV) ions crystallize as sulphates at low temperatures, the operation temperature range is limited to 10-40°C (Wang et al., 2018; Xi et al., 2016). Further limitations on energy density include the anode electrolyte's instability (Rahman & Skyllas-Kazacos, 2009), membrane cross-contamination, and the water electrolysis-induced limitation on cell potential in aqueous systems (Gores & Barthel, 1983).

Because of their affordability and compatibility with vanadium ions, sulphate-based electrolytes are extensively employed in vanadium redox flow batteries (VRFBs). Because VRFBs rely on redox processes, the electrolyte is usually a sulfuric acid solution of vanadium sulphate. This solution may take on a variety of oxidation states, including V(II), V(III), V(IV), and V(V) (Kausar et al., 2016).

3.2.2.1 Concentration of electrolyte

Wang et al. (2018) studied the effect of changing the concentration of vanadium and sulfuric acid on the stability of the electrolyte over a broad range of temperature (238.15K-333.15K). The VO_2^+ electrolytes, including diverse vanadium concentrations (0.4 M-2.0 M) and variable sulfuric acid concentrations (1.5 M-3.0 M), were synthesized by dissolving vanadyl sulphate ($VOSO_4 \cdot 3.5H_2O$, 99% purity) in concentrated sulfuric acid solutions. The electrolytes, classified into four valence states (V^{2+}, V^{3+}, VO^{2+}, and VO_2^+), were analyzed using static temperature stability tests performed in a thermostat with a range of -35°C to 60°C. It was found that increasing vanadium concentrations enhanced energy density but diminished temperature stability, with peak performance obtained at 1.4 to 1.6 M vanadium and 2.0 to 2.5 M sulfuric acid, achieving an equilibrium between energy density and thermal

adaptability. The VFBs demonstrated consistent performance throughout a temperature range of -25°C to 60°C without additives, signifying a notable technical development. At low temperatures, performance was limited by elevated polarization resistance, but at higher temperatures, vanadium ion permeability reduced Coulombic efficiency but improved voltage efficiency.

The supersaturated 3M vanadium electrolyte to enhance the energy density. Previously, it was observed that vanadium ions precipitate at high temperature and concentration, so many additives are used as stabilizers in this research. Different phosphates and sulphates are prepared with the required concentration and used as additives. The research indicated that the incorporation of 1 wt% H_3PO_4 with 2 wt% ammonium sulfates significantly enhanced the stability of the 3 M vanadium electrolyte, postponing precipitation. This combination exhibited enhanced stability at elevated temperatures, sustaining V(V) concentrations over 2.5 M for prolonged durations. Electrochemical experiments demonstrated that the improved electrolyte composition facilitated stable cycling performance, achieving coulombic efficiencies of about 90% and maintaining energy efficiency stability beyond 90 cycles. This 3 M electrolyte composition enhanced energy density by 60–90% compared to standard 1.6 M solutions.

In another study, Yang et al. (2020) studied the effect of state of charge (SOC) on energy density and electrolyte stability. V(IV) electrolytes were prepared by the electrolytic dissolution of V_2O_5 in sulfuric acid, resulting in the formation of V(V), V(III), and V(II) electrolytes via oxidation or reduction processes. The research indicated that a vanadium electrolyte concentration of 2.0 M and a sulfate concentration of 5.5 M exhibited stability from -10°C to 40°C when the state of charge (SOC) ranged from 0% to 90%, with stability improving as SOC decreased owing to complexation interactions. Increased vanadium concentrations improved energy density, while variations in viscosity and conductivity affected performance. Electrochemical activity was maximized at 0% and 100% state of charge (SOC) but diminished at 50% SOC owing to polarization and suboptimal kinetics. Spectral analysis indicated that the synthesis of $[V_2O_3(H_2O)_7]^{3+}$ enhanced thermal stability, while decreasing electrochemical efficiency. In general, VRFBs demonstrated stable energy efficiency (75–80%) over the temperature spectrum.

3.2.2.2 Addition of additives

Another way to increase the energy efficiency as well as energy density of vanadium redox flow batteries is by the addition of additives in the electrolyte, as shown in the table. By the addition of additives, stability of electrolyte increases as well as electrochemical performance also improved a study by Gang Wang shows that addition of various additives like methyl orange (MO), Triton X-100 (OP), sodium Ligninsulfonate (SL), sodium dodecyl sulfate (SDS), and polyvinyl alcohol (PVA) increases the stability of electrolyte even at high concentration exceeding 2 M. Other than stability, this addition enhances the energy capacity of VRFBS by increasing the retention of higher vanadium concentrations for an extended period (Wang et al., 2013a). Despite that, these additives are temperature dependent and less stable compared to some other like HMTA (Hexamethylenetetramine) and EDTADS (Ethylenediaminetetraacetic Acid Disodium Salt).

To overcome problems related to thermal stability at elevated temperatures, Zhangxing He used methanesulfonic acid (MSA) and Aminomethylsulfonic acid (AMSA) as an additive. The VRFB cell with AMSA demonstrated enhanced cycling performance, attaining greater energy efficiency (81.5%) and better capacity retention over 40 cycles relative to the pristine electrolyte. XPS study indicated that AMSA incorporated NH_2 and SO_3H groups into the graphite felt surface, increasing hydrophilicity and offering more active sites for electrochemical processes, thereby improving electrode performance (He et al., 2013).

Table 1. Effect of different additives on the electrochemical performance of VRFBS

Study	Additives used	Key findings
Wang et al. (2013a)	Methyl Orange (MO), Triton X-100 (OP), Sodium Ligninsulfonate (SL), Sodium Dodecyl Sulfate (SDS), Polyvinyl Alcohol (PVA)	Enhanced stability of vanadium electrolyte solutions (>2.0 M); increased charge transfer efficiency; decreased polarization resistance; elevated energy density.
He et al. (2013)	Methanesulfonic Acid (MSA), Aminomethylsulfonic Acid (AMSA)	Improved thermal stability of V(V) electrolyte; enhanced cycling performance; greater energy efficiency (81.5%); better capacity retention.
Wang et al. (2014) (Acid Additives)	Methanesulfonic Acid, Boric Acid, Hydrochloric Acid, Trifluoroacetic Acid, Polyacrylic Acid, Phosphotungstic Acid	Improved thermal stability, elevated vanadium concentrations, inhibited precipitation, and enhanced energy density across temperatures.
Yang et al. (2020)	Taurine	Improved energy efficiency, stable cycling performance, enhanced thermal stability.
Lei et al. (2013)	L-Glutamate	Enhanced thermal stability, increased peak currents, decreased polarization resistance, improved energy density.

Other studies by Gang Wang using acid additives such as methanesulfonic acid, boric acid, hydrochloric acid, trifluoroacetic acid, polyacrylic acid, and Phosphotungstic acid (Wang et al., 2014), by Yang et al. (2020) using taurine (Yang et al., 2018). While Lei et al. (2013) used amino acid as an additive and observed the stability of the electrode at elevated temperature also showed the same results. Researchers discovered that adding different chemicals, like methanesulfonic acid, boric acid, taurine, and L-glutamate, made vanadium redox flow batteries (VRFBs) much better at storing energy and working overall. Those modifications improved the thermal stability of the V(V) electrolyte, preserving elevated vanadium concentrations and inhibiting precipitation, which is essential for sustaining energy density across different temperatures. Enhanced electrochemical performance was shown by elevated peak currents, decreased polarization resistance, and augmented diffusion coefficients, all of which promote efficient redox processes and enhance energy density. The VRFB cells using optimal additives, including taurine and L-glutamate, demonstrated superior energy efficiency and consistent cycling performance, highlighting the significance of these additives in improving energy storage and sustaining successful charge-discharge cycles.

3.2.2.3 New redox pair

Wu et al. (2014) also developed and examined an innovative high-energy-density positive electrolyte for redox flow batteries (RFBs) that combines various redox couples, especially the Fe^{3+}/Fe^{2+} and Br_2/Br^- couples. It was observed that a mixed electrolyte consisting of Fe^{3+}/Fe^{2+} and Br_2/Br^- attained an impressive energy density of 827 Wh/L and a capacity density of 804 Ah/L, which much exceeds the performance of standard electrolytes. Both redox couples exhibited remarkable reversibility on graphite electrodes, with Br^- significantly improving the reversibility of the Fe^{3+}/Fe^{2+} pair. Furthermore, the flow cell exhibited consistent charge/discharge performance, shown by two separate voltage plateaus for the Fe^{3+}/Fe^{2+} and Br_2/Br^- redox couples, confirming the electrolyte's efficient energy storage properties. Despite these, enhanced viscosity and ionic interactions at greater concentrations might hinder performance. Although increased temperatures enhance reaction kinetics, they also aggravate problems like electrolyte instability and electrode deterioration over time, highlighting the need for ongoing research.

3.2.2.4 Other methods

To enhance the energy density of non-aqueous vanadium redox flow batteries, T. Herr, P. Fischer, and colleagues formulated a solvent combination including acetonitrile, dimethyl sulfoxide, and 1,3-dioxolane while using tetrabutylammonium hexafluorophosphate as a supporting electrolyte. It was observed that the addition of a solvent mixture enhances the stability of vanadium acetylacetonate redox flow batteries, hence increasing the energy density. On the other hand, the addition of a small amount of dimethyl sulfoxide increases the electrolyte conductivity, but excessive addition increases the viscosity and hence decreases the conductivity. Other than that, an energy efficiency of 27% while a coulombic efficiency of 95% is observed. Despite these, high ohmic losses as well as formation of by-products like vanadyl acetylacetonate ($VO(acac)_2$) cause capacity degradation with the passage of time (Herr et al., 2014).

In another study, Yang et al. (2019) used a sulfate and chloride mixed acid electrolyte and studied the behavior and electrochemical process at a broad temperature range of -20°C to 50°C. Electrolytes with varied concentrations of vanadium, sulfate, and chloride were prepared and optimal stability was attained with a vanadium concentration of 2.4M, a chloride concentration ranging from 6.0 to 6.4M, and a sulphate concentration between 2.0 and 3.0M, enabling VRFBs to operate across a temperature range of -20°C to 50°C, contributing to high energy density. Chloride ions provide stable complexes that prevent precipitation at high temperatures, improving V(V) thermal stability. Sulphate and chloride ions make lower-valence vanadium ions soluble at low temperatures, inhibiting crystallization. The mixed acid's high acidity prevents precipitation at different temperatures, while chloride ion concentrations at 6.4M minimize hydrogen chloride volatilization, ensuring stable electrolyte conditions.

To summarize, enhancing the energy density of vanadium redox flow batteries (VRFBs) requires strategic changes to the electrolyte composition and operating conditions. Optimizing vanadium and acid concentrations, adding stabilizing additives such as taurine, AMSA, and phosphate-based compounds, and using mixed acid systems with sulfate and chloride ions have all been shown to improve thermal stability, prevent precipitation, and maintain electrochemical efficiency across a wider temperature range. Advanced electrolyte formulations, such as those including methanesulfonic acid or solvent mixes like acetonitrile and dimethyl sulfoxide, reveal additional promise by balancing conductivity, stability, and viscosity. These developments help to increase energy densities while addressing issues such as limited temperature stability, polarization resistance, and electrode deterioration. Future studies should concentrate on fine-tuning these improvements and investigating novel redox combinations to improve VRFB performance for sustainable and efficient energy storage options.

3.3 Membrane

Membranes are essential for the effectiveness and economic feasibility of VRFBs, as they separate the two half-cells and facilitate ion exchange while minimizing cross-contamination. Polymer electrolyte membranes not only provide physical separation of the electrolytes, but they also help to maintain the overall system's operating efficiency by preventing short circuits and reducing the crossover of unwanted active species (Lu et al., 2017). Although they are critical components, membranes are among the most expensive VRFB components, accounting for a sizeable portion of the overall system's capital cost (Zeng et al., 2015). Cation-exchange, anion-exchange (Couture et al., 2011), amphoteric-ion exchange, and porous membranes (Lu et al., 2017) are examples of membranes used to improve the ion conductivity, selectivity, and mechanical stability of VRFBs. However, high vanadium permeability, reduced voltage efficiency at high current densities, and trade-offs between ionic conductivity and selectivity necessitate further research in this field. Recent advances in membrane material composition, structural modification, and membrane

thickness optimization have shown promise for increasing VRFB energy density, operational efficiency, and longevity (Chen et al., 2013b).

Hwang & Ohya (1996) synthesized cation exchange membrane by chlorosulfonated polyethylene as a separator in VRFBS. Vapor-phase chlorosulfonation is used to synthesized asymmetric membrane. High energy efficiency of 85% is observed while this also shows lower vanadium ion permeability as compared to other. In another study he also synthesized cross-linked anion exchanges membrane through accelerated electron radiation. Current density of 93.5%, voltage efficiency of 87.7% while overall efficiency of 82% is observed which is quite high as compared to non-crosslinked membrane (Hwang & Ohya, 1997).

3.3.1 Type of membranes

3.3.1.1 Perfluorinated ion-exchange membranes

Perfluorosulfonic acid (PFSA)-based cation-exchange membranes, particularly the Nafion series, have long been the benchmark separators in vanadium redox flow batteries (VRFBs) due to their high proton conductivity, chemical robustness, and mechanical stability under acidic and oxidative conditions (He et al., 2025). Their perfluorinated backbone provides strong C–F bonds that resist degradation during prolonged cycling, ensuring reliable long-term operation. However, the negatively charged sulfonic matrix of PFSA membranes facilitates the diffusion of cationic vanadium species (V^{2+}, V^{3+}, VO^{2+}, VO_2^+), leading to capacity decay and efficiency loss through cross-contamination between half-cells (Huynh et al., 2025).

Efforts to mitigate vanadium crossover in Nafion have focused on structural and compositional modifications. Vrána et al. demonstrated that ion-exchange capacity (IEC) and internal morphology critically influence vanadium permeability, with higher IEC improving proton conduction but also enhancing vanadium ion diffusion(Vrána et al., 2018). To address this trade-off, Moon et al. introduced pore-filled fluorinated ion-exchange membranes using a porous polyethylene reinforcement, which improved oxidative stability and dimensional control without compromising conductivity (Moon et al., 2021).

Recent advances have explored partially fluorinated hybrid systems to balance performance and cost. For instance, Sharma and Shahi developed a partially fluorinated sulfonated polymer hybridized with MIL-101(Cr), achieving 97.5% coulombic efficiency at 150 mA cm^{-2} and stability over 1500 cycles (≈650 h). The improvement was attributed to synergistic interactions between Cr-MOF and the fluorinated matrix, forming hydrophilic proton-conducting channels that enhanced selectivity while reducing crossover. Such partially fluorinated hybrids offer a cost-effective alternative to fully fluorinated Nafion membranes while retaining their chemical stability (Sharma & Shahi, 2023).

Although perfluorinated membranes such as Nafion remain widely used, their high cost and vanadium permeability continue to limit large-scale deployment. Therefore, current research increasingly favors hybrid, reinforced, or partially fluorinated membranes that preserve the durability of PFSA structures but incorporate tailored morphologies or fillers to suppress vanadium diffusion and enhance overall VRFB energy efficiency.

3.3.1.2 Hydrocarbon-based ion-exchange membranes

Hydrocarbon-based ion-exchange membranes have emerged as promising alternatives to costly perfluorinated membranes (e.g., Nafion) for vanadium redox flow batteries (VRFBs) due to their lower cost, tunable structure, and high proton selectivity. Unlike perfluorinated systems, these membranes possess smaller ionic clusters that restrict vanadium ion crossover while maintaining adequate proton conductivity, thereby improving coulombic efficiency (CE) and overall energy efficiency (EE) (Wang et al., 2020).

Yu *et al.* (2018) demonstrated that integrating a hydrocarbon membrane, specifically sulfonated poly(ether ketone) (SPEEK), with $Ce_{0.8}Zr_{0.2}O_2$-modified graphite felt (GF)

electrodes significantly enhanced VRFB performance compared to conventional Nafion-based systems. The SPEEK membrane effectively suppressed vanadium crossover, yielding higher CE and cycling stability, while the $CexZr_{1-x}O_2$ catalyst improved electrode wettability and electrochemical activity toward VO_2^+/VO^{2+} and V^{2+}/V^{3+} redox couples. This combination achieved superior voltage and energy efficiencies at a markedly reduced material cost, demonstrating the potential of hydrocarbon membranes for practical large-scale applications.

Building on these advantages, Wang *et al.* developed a new class of sulfonated aromatic polymer membranes designed to overcome the intrinsic trade-off between proton conductivity and ion selectivity. Their optimized polymer, BP-ArF_4, contained a biphenyl backbone with perfluorinated aromatic side chains, which established a well-defined hydrophilic–hydrophobic phase separation and strong hydrogen bonding with vanadium species. This structure produced narrowly distributed ionic domains (<7 Å), achieving exceptional proton/vanadium selectivity. As a result, VRFBs employing BP-ArF_4 exhibited a coulombic efficiency of ~99% and energy efficiency up to 93.9%, surpassing all conventional IEMs (Wang et al., 2022).

Overall, hydrocarbon-based membranes such as SPEEK and sulfonated aromatic polymers demonstrate that careful molecular design, optimizing ionic domain size, functional group distribution, and polymer backbone composition, can deliver low-cost, chemically stable, and highly selective membranes. These developments are key to advancing next-generation VRFBs for grid-scale energy storage.

3.3.1.3 Anion-exchange membranes (AEMs)

Anion-exchange membranes (AEMs) have emerged as promising alternatives to conventional cation-exchange membranes for vanadium redox flow batteries (VRFBs), due to their ability to reduce vanadium ion crossover, enhance Coulombic efficiency, and extend battery life. AEMs contain positively charged functional groups, such as quaternary ammonium, imidazolium, or piperidinium, which generate an electrostatic field that repels vanadium ions through the Donnan exclusion effect, thereby limiting electrolyte cross-contamination and reducing the frequency of electrolyte rebalancing (Chen et al., 2013a; Lallo et al., 2022). Various types of AEMs have been explored, including quaternary ammonium-functionalized aromatic polymers, which exhibit tunable ion exchange capacities (IEC) and vanadium selectivity. Chen et al. demonstrated that QA-Radel membranes with an IEC of 2.0 mequiv g^{-1} achieved a balance between ionic conductivity and vanadium retention, delivering higher maximum power densities than Nafion N212, while lower IEC membranes showed higher Coulombic efficiency but reduced voltage efficiency (Chen et al., 2013a). Commercial imidazolium-functionalized Aemion™ membranes have also shown excellent chemical stability and low vanadium permeability, maintaining performance over extended cycling with minimal structural degradation (Lallo et al., 2022).

Cardo-polyetherketone (PEK-C) based AEMs quaternized with trimethylamine exhibited sulfate ion conductivities of 5.6 mS cm^{-1} and vanadium(IV) permeability of 8.2 × 10^{-9} cm^2 s^{-1}, retaining mechanical integrity and chemical stability during prolonged exposure to VO^{2+} solutions, with VRFB testing demonstrating Coulombic and energy efficiencies of 98% and 80%, respectively (Yun et al., 2014). Redox-active membranes incorporating ferrocene moieties have been developed to further enhance ion selectivity and prevent vanadium crossover. These membranes, synthesized via free radical polymerization of tert-butyl methacrylate, vinylimidazole, and vinyl ferrocene, exploit the redox activity of ferrocenium/ferrocene centers to improve ionic transport and chemical robustness, achieving 98% Coulombic efficiency and 84.5% energy efficiency over 150 cycles (Sharma et al., 2021). Similarly, long side-chain quaternary ammonium-functionalized poly(arylene piperidinium) membranes display exceptionally low vanadium permeability, high H^+ conductivity, and ion selectivity up to 370 times higher than Nafion 115, resulting in Coulombic efficiencies above 99% and energy efficiencies up to 89.8%

under typical operating conditions (Che et al., 2022). Bifunctional cross-linked fluorinated poly(aryl ether oxadiazole) membranes, using poly(N-vinylimidazole) as a cross-linker, have demonstrated high ion exchange capacities, reduced vanadium permeability, and water uptake of 68 wt%, providing chemical stability in strongly acidic VO^{2+} solutions and achieving Coulombic and energy efficiencies greater than 92% and 86%, respectively, over multiple cycles (Wang et al., 2020).

3.3.1.4 Composite and hybrid membranes

Composite and hybrid membranes have emerged as a vital direction in advancing vanadium redox flow batteries (VRFBs), addressing the limitations of conventional perfluorinated and hydrocarbon membranes. By integrating inorganic fillers or secondary polymers into ionomer matrices, these membranes exhibit enhanced mechanical strength, chemical durability, and reduced vanadium ion crossover, key factors for extending VRFB lifetime and efficiency.

Early studies on Nafion–TiO_2, Nafion–SiO_2, and ZrP–SPEEK composites demonstrated the synergistic effects of inorganic–organic interfaces in improving oxidative stability and ion selectivity (Di et al., 2021; Zhang et al., 2021). The inclusion of oxides such as TiO_2 and SiO_2 introduces tortuous diffusion paths that hinder VO_2^+ ion transport while maintaining proton conductivity through connected hydrophilic channels. Likewise, ZrP-modified sulfonated poly(ether ketone) (SPEEK) membranes displayed superior chemical resistance and dimensional stability under acidic conditions due to strong acid–base interactions between the filler and polymer backbone (Wang et al., 2020).

The incorporation of graphene oxide (GO) into sulfonated polymers (e.g., SPEEK/GO) further enhanced ion selectivity by leveraging GO's layered structure and oxygen functional groups, which form hydrogen-bond networks facilitating proton conduction while blocking vanadium ion crossover (Pahlevaninezhad et al., 2023). Additionally, PVDF-supported hybrid systems offered mechanical reinforcement and dimensional control, combining hydrophobic PVDF layers with sulfonated polymer matrices to minimize swelling and preserve conductivity during long-term cycling (Ren et al., 2017).

Significant advancement occurred with perfluorinated hybrid membranes, especially through the modification of Nafion structures. Sadhasivam and colleagues introduced an alkoxysilane-functionalized polymer (ASFP) into Nafion to form a Nafion–ASFP hybrid membrane through covalent interconnection of hydrophilic domains. The optimized Nafion–ASFP (75:25) membrane achieved a proton conductivity of 0.061 S cm^{-1} and low vanadium permeability (1.26×10^{-7} cm^2 min^{-1}), maintaining stable performance over 50 cycles with higher Coulombic efficiency than Nafion 212 (Sadhasivam et al., 2017). This design demonstrated how introducing an inorganic–organic precursor polymer can improve both selectivity and cost-effectiveness. Complementary findings from SiO_2-doped perfluorinated systems further confirmed that uniform filler dispersion within the fluoropolymer matrix produces consistent phase separation, balancing mechanical strength and ionic conductivity (Ling et al., 2019). Such hybridization strategies effectively overcome the conductivity–selectivity trade-off common in single-phase ionomers.

Recently, Liu et al. developed Aquivion-based hybrid membranes by incorporating a norbornene-derived polymer synthesized through ring-opening metathesis polymerization. The hybrid (Aquivion/PAN-2.5%) membrane achieved strong "acid–base" hydrogen bonding and ionic crosslinking, leading to lower VO_2^+ permeability and improved Coulombic and voltage efficiencies. The system maintained over 0.8 V for 31 h and attained an energy efficiency of 73.02% at 160 mA cm^{-2} (Liu et al., 2021). These results highlight how molecular-level hybridization and short-sidechain perfluorinated ionomers can simultaneously optimize proton selectivity and long-term durability.

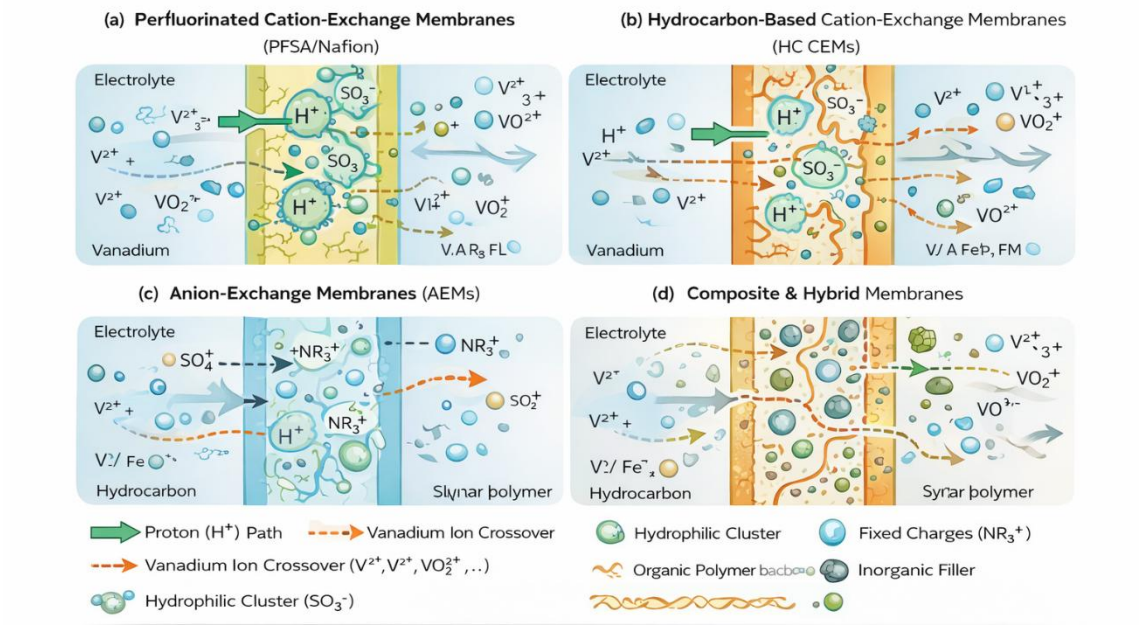

Fig. 5. Schematic comparison of membrane types in vanadium redox flow batteries

Figure 5 provides a comparative schematic of the major membrane classes employed in vanadium redox flow batteries, highlighting their distinct ion-transport mechanisms and structure–property relationships. Perfluorinated cation-exchange membranes (PFSA/Nafion) offer high proton conductivity through interconnected sulfonic acid clusters but suffer from significant vanadium ion crossover. Hydrocarbon-based cation-exchange membranes feature smaller ionic domains that improve vanadium selectivity while maintaining sufficient proton transport at lower cost. Anion-exchange membranes suppress vanadium crossover via Donnan exclusion arising from fixed positive charges, enhancing Coulombic efficiency but often at the expense of conductivity. Composite and hybrid membranes combine polymer matrices with inorganic fillers or reinforcements to create tortuous diffusion pathways, effectively reducing vanadium permeability while preserving proton-conducting channels. Overall, the figure summarizes how membrane design strategies aim to balance conductivity, selectivity, durability, and cost to improve long-term VRFB performance.

3.3.2 Modification in membrane to enhance energy density

A lot of research has been done to increase the energy efficiency and density by modifying the surface of these membranes of VRFBS. S.M. Hosseini and colleagues did the surface modification of a cation exchange membrane by coating the membrane surface with silver nanoparticles in a vacuum reactor using argon plasma treatment. It was found that maximum energy efficiency was calculated using a 40nm thick silver nanoparticle coating. Low electrical resistance (4.66 $\Omega \cdot cm^2$ compared to 11.54 $\Omega \cdot cm^2$ for the unmodified membrane), maximum current efficiency (98.01%), and reduced energy consumption were observed by this modification, which increases the overall energy density as compared to the unmodified one (Hosseini et al., 2010).

In another study by Akbar Zendehnam and colleagues, the heterogeneous modification of CEMs with a matrix blend of polyvinyl chloride and styrene-butadiene rubber has been performed by adding Fe-Ni oxide nanoparticles and an Ag nanolayer. An improvement in electrical conductivity, membrane potential, perm selectivity, and ion flux is observed by modification using 10-20nm thick silver layer. This improved perm selectivity and high ion-flux indicate an increase in energy efficiency and density (Zendehnam et al., 2013). Despite these, high temperature requirements, high-cost

equipment, and energy consumption needed to maintain high temperatures are the drawbacks of the plasma modification method.

To overcome this challenge, electrodeposition is used for surface modification of the membrane. Jie Zeng and colleagues compared the effectiveness of electrolyte soaking, oxidation polymerization, and electrodeposition. Their study found that Pyrrole monomers deposited on Nafion membranes shows reduced in vanadium ion permeability (2.87×10^{-6}cm^{2}min^{-1} (untreated Nafion) to 5.0×10^{-7} cm^2 min^{-1}), which shows reduced self-discharging, increased energy efficiency, and density. Other than that, the proton conductivity of electrodeposited membranes also increased, reaching 7.83×10^{-3} S/cm, higher than other modification methods (Zeng et al., 2008).

In another research, pores in the membrane are filled, and then their impact on the electrochemical process of VRFBS was studied. For this, Yeonho Ahn and Dukjoon Kim hydrophilize hydrophobic PTFE substrates with catechol and polyethyleneimine (PEI) to allow better impregnation. To this end, the imidazolium grafted poly (arylene ether ketone) (PAPI) was synthesized and filled into the pores of PTFE to form the reinforced functional membrane. PTFE/PAPI membranes achieved significantly lower vanadium ion permeability (1.4×10^{-7} to 2.08×10^{-7} cm^2 min^{-1}), which is 10 times lower than Nafion 117 and 3 times lower than FAP450. Despite the presence of a non-conductive PTFE substrate, ionic conductivity was enhanced and was comparable to commercial membranes like FAP450. Energy efficiency also increased as compared to other and hence energy density increases too (Ahn & Kim, 2022). Overall, the comparison of modification techniques is shown in table 2.

Table 2. Comparison of different surface modification techniques used for VRFBS membrane

Surface modification technique	Material used	Key outcomes	Challenges
Plasma Coating	Ag nanoparticles (40 nm)	Low resistance (4.66 Ω·cm^2); 98.01% current efficiency	Elevated temperature requirements
	Ag nanolayer (20nm)	improved electrical conductivity, membrane potential, perm selectivity, and ion flux	Elevated temperature requirement
Electrodeposition	Pyrrole monomers on Nafion membranes	Reduced vanadium permeability; 7.83×10^{-3} S/cm proton conductivity	Scalability issues
Pore-Filling	PTFE + PAPI	Vanadium permeability 10× lower than Nafion; high energy efficiency	Complex preparation

In addition to surface modification, extensive research has been conducted to enhance the energy density and efficiency of these systems. For instance, Dasom Jeong and colleagues developed a novel composite membrane incorporating hydrophilic carbon quantum dots (CQDs) synthesized from human hair via the hydrothermal method. The incorporation of CQDs significantly improved the membrane's performance, increasing the coulombic efficiency to 96.13% and the energy efficiency to 88.2%. These enhancements were attributed to the increased proton conductivity, reduced charge crossover, lower vanadium ion permeability, and decreased self-discharge rate, all of which contribute to achieving a higher energy density in the batteries (Jeong et al., 2024). Guihui Xie and colleagues prepared free-standing COF nanofiber mats using electrospinning and densified with sulfonated polybenzimidazole (SPBI) to create continuous ion-conductive pathways. The COF fiber ICM with 15% nanofiber content achieved an energy efficiency of 80.5%, significantly outperforming commercial Nafion 212 (74.9%) and pristine SPBI membranes (77.5%). Other than that, these membranes show 3 times more ion selectivity and much lower self-discharging than Nafion 212 and pristine SPBI membranes, showing high energy density (Xie et al., 2024).

In conclusion, membrane modification techniques have significantly improved VRFB performance, particularly in terms of energy density and operational efficiency. Surface coatings with silver nanoparticles and pore-filling with functional polymers like PAPI have shown significant improvements in reducing vanadium ion permeability, increasing ion selectivity, and increasing proton conductivity. Electrodeposition and the incorporation of novel materials, such as carbon quantum dots and COF nanofibers, have also helped to achieve higher energy efficiencies, often outperforming traditional membranes like Nafion. However, high-cost equipment, complex preparation processes, and scalability remain obstacles to widespread adoption.

3.4 Future recommendation

Despite significant advancements in electrode engineering, electrolyte optimization, and membrane modification, Vanadium Redox Flow Batteries (VRFBs) still face fundamental challenges that limit their energy density and large-scale applicability. These include restricted electrolyte solubility, low cell voltage, sluggish redox kinetics, and cost constraints. Therefore, future research should focus on multi-dimensional strategies that integrate material innovation, system optimization, and sustainability-driven design to further enhance the energy density and overall performance of VRFBs.

3.4.1 Advanced electrode design and catalytic engineering

Future work should emphasize rational electrode architecture that simultaneously improves electrochemical activity, mass transport, and structural durability. Emerging fabrication techniques such as atomic layer deposition (ALD), electrospinning, and 3D printing can enable precise control over electrode morphology and porosity, thus reducing polarization losses and enhancing current distribution (Kim et al., 2015). The development of heteroatom-doped carbon materials (N, S, or B doping), MXene-based composites, and high-entropy alloy oxides (HEAOs) presents a promising route for improving redox kinetics of both the V^{2+}/V^{3+} and $VO_2{}^{+}/VO_2{}^{++}$ couples (Agarwal et al., 2024)

In addition, multi-element catalytic systems combining transition metals (e.g., Nb, Mo, and W) with conductive carbon frameworks can enhance surface reactivity and suppress the hydrogen evolution reaction, thereby improving energy efficiency (Gencten & Sahin, 2020). Integrating machine learning (ML)-assisted modelling to predict optimal electrode configurations and simulate mass transfer behaviors could accelerate the rational design of next-generation electrodes for high-power VRFBs (Olabi et al., 2023).

3.4.2 Electrolyte optimization and redox chemistry development

Electrolyte formulation plays a decisive role in determining the energy density and operational window of VRFBs. Future research should focus on developing highly concentrated, thermally stable, and wide-temperature-range electrolytes that maintain stability across −20°C to 60°C. Mixed-acid systems such as sulfate, chloride, and methanesulfonic acid (MSA) blends have demonstrated enhanced vanadium solubility and reduced V_2O_5 precipitation at elevated temperatures (Tian et al., 2023). The incorporation of organic additives such as taurine, L-glutamate, and aminomethylsulfonic acid (AMSA) has been shown to improve the thermal and electrochemical stability of V(V) electrolytes while increasing coulombic and energy efficiency (Düerkop et al., 2021)

Further progress may arise from exploring non-aqueous or hybrid aqueous–organic electrolytes employing solvents such as dimethyl sulfoxide (DMSO), acetonitrile, and ionic liquids, which could expand the operating voltage beyond 1.6 V and increase theoretical energy density (Yang et al., 2024). Developing self-regulating electrolytes capable of autonomously correcting redox imbalances through reversible complexation or dynamic pH buffering would further stabilize long-term cycling (Yu et al., 2024).

3.4.3 Membrane innovation and functional hybridization

The membrane remains one of the most influential components affecting coulombic efficiency, voltage stability, and overall system cost. Therefore, future studies should target the trade-off between proton conductivity and vanadium ion permeability by introducing functional hybrid membranes that combine the advantages of perfluorinated and hydrocarbon polymers. Incorporating inorganic fillers such as TiO_2, SiO_2, ZrP, and graphene oxide (GO) into polymer matrices has proven effective in creating tortuous ion diffusion paths that suppress vanadium crossover while maintaining high proton conductivity (Palanisamy & Oh, 2022).

Emerging materials such as Covalent Organic Frameworks (COFs) and Metal–Organic Frameworks (MOFs), due to their tunable pore sizes and high chemical stability, offer a new platform for achieving selective ion transport and long-term durability (Yu et al., 2024). Similarly, bio-derived nanomaterials such as carbon quantum dots (CQDs) synthesized from sustainable precursors (e.g., human hair or cellulose) have demonstrated superior hydrophilicity and ionic selectivity, leading to enhanced coulombic and energy efficiencies exceeding 96% and 88%, respectively (Jeong et al., 2024). Moreover, pore-filling composite membranes integrating poly(arylene ether ketone) (PAPI) or sulfonated polybenzimidazole (SPBI) into PTFE substrates can further lower vanadium permeability by an order of magnitude while maintaining mechanical robustness (Ahn & Kim, 2022). Future investigations should also include in-situ membrane characterization under realistic VRFB conditions to monitor degradation and fouling, allowing real-time assessment of chemical and mechanical stability (Guarnieri et al., 2016).

3.4.4 System-level optimization and stack engineering

Beyond materials development, system-level optimization offers substantial potential for enhancing energy density. Advanced flow field geometries, such as interdigitated or serpentine designs, can promote uniform electrolyte distribution, minimize dead zones, and improve mass transfer at high current densities (Skyllas-Kazacos et al., 2011). Moreover, designing compact and lightweight stack architectures using conductive polymer composites or metal–polymer hybrid bipolar plates can reduce system volume and improve energy density per unit mass (Satola, 2021).

Integrated thermal management systems are also essential to maintain electrolyte stability, especially under high-current operations or variable environmental conditions. Employing computational fluid dynamics (CFD) and multiphysics simulations can aid in optimizing flow uniformity, minimizing pressure drop, and predicting performance under dynamic grid conditions (Wang et al., 2023). Incorporating hybrid energy storage configurations, such as coupling VRFBs with supercapacitors or hydrogen fuel cells, can address both short- and long-duration storage needs, improving system flexibility and grid reliability (Schubert et al., 2023).

3.4.5 Economic viability and sustainability considerations

To ensure the long-term adoption of VRFBs, it is crucial to align performance improvements with economic and environmental sustainability, and future work should prioritize the development of low-cost hydrocarbon-based membranes to replace expensive perfluorinated polymers like Nafion, without compromising stability (Düerkop et al., 2021), the implementation of closed-loop vanadium recovery and electrolyte recycling processes, which can reduce raw material dependency and minimize environmental impact (Blume et al., 2024), and the exploration of green synthesis pathways using biomass precursors for carbon-based electrodes and membranes to lower carbon footprint and enhance sustainability. Conducting life cycle assessments (LCA) and techno-economic analyses (TEA) of newly developed materials and configurations will provide a clearer understanding of their scalability and long-term feasibility.

4. Conclusions

This review highlights the major advancements in enhancing the energy density of Vanadium Redox Flow Batteries (VRFBs) through innovations in electrodes, electrolytes, and membranes. While VRFBs are already recognized for durability and scalability, their low energy density remains a core limitation. Research on electrode surface modification, electrolyte optimization, and membrane functionalization has significantly improved electrochemical performance, efficiency, and long-term stability, moving VRFBs closer to commercial large-scale energy storage deployment. Electrode engineering, through nano-structuring, catalytic coatings, and multi-metal oxide doping, has accelerated redox kinetics and reduced side reactions such as hydrogen evolution, boosting energy efficiency.

Optimized electrolyte formulations and stabilizing additives have expanded temperature ranges and improved stability, while hybrid organic–inorganic membranes have minimized vanadium crossover and enhanced proton selectivity. At the system level, innovations in stack design, thermal management, and computational modelling are improving compactness and cost-effectiveness. These developments demonstrate that VRFB optimization is inherently interdisciplinary, integrating materials science, electrochemistry, and system engineering. Overall, these advancements contribute to the broader goal of sustainable, efficient, and long-duration energy storage essential for renewable energy integration. Future research should emphasize scalable synthesis of advanced materials, low-cost hydrocarbon membranes, closed-loop electrolyte recycling, and hybrid storage systems combining VRFBs with supercapacitors or hydrogen fuel cells. With continued innovation, VRFBs are well-positioned to become a cornerstone of a clean and resilient energy infrastructure.

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During the preparation of this work, the author(s) used a generative AI tool to assist in paraphrasing certain sections for clarity, Grammarly to assist in improving the grammar and academic tone of the manuscript, and generative AI tools for figure generation. After

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