

Article

Quantitative Solid-Liquid Matching and Capacity Optimization of Prussian Blue Redox Targeting in Neutral Iron-Sulfur Flow Batteries

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Abstract: Neutral iron-sulfur flow batteries feature low cost, high safety and environmental friendliness, demonstrating great application potential for large-scale energy storage. However, their cathodic capacity is fundamentally limited by the low solubility of ferricyanide active species. Most previous works merely verified the reversible redox reaction between Prussian blue (PB) and ferrocyanide in symmetric single-electrolyte cells, without systematic quantitative research on solid-liquid matching mechanisms and practical full-cell performance evaluation in iron-sulfur systems. This study explores cathode capacity enhancement and full-cell electrochemical characterization to solve the above bottleneck. Nickel sulfide modified graphite felts were synthesized via an in-situ hydrothermal route, and a redox-targeted symmetric cell was assembled with ferricyanide mediators and solid PB. CV results reveal favorable potential matching, while XRD measurements confirm the reversible phase transition between PB and Prussian white (PW) during cycling. The 0.3 M ferricyanide electrolyte achieves effective PB activation, and among 10–40% addition ratios, the 20% PB loading delivers the optimal capacity increment and material utilization, yielding a maximum discharge capacity of 69.66 mAh and a peak power density of 14.58 mW cm⁻² in full-cell tests. Integrating 20% PB into full cells remarkably elevates coulombic, voltage and energy efficiencies, with voltage efficiency improved most prominently. Energy efficiency drops continuously as current density rises from 20 to 60 mA·cm⁻². The battery reaches its maximum energy efficiency at 40 °C, since mild heating accelerates solid-liquid redox targeting kinetics without inducing PB lattice distortion or ferrocyanide hydrolysis.

Keywords: neutral iron-sulfur flow battery; Prussian blue; redox-targeted reaction; energy efficiency

1. Introduction

Driven by continuous socioeconomic development, global energy demand has experienced a steady increase. The long-term large-scale exploitation and utilization of traditional fossil fuels have exacerbated resource constraints, environmental contamination, and greenhouse gas emissions, posing severe challenges to energy security and ecological conservation [1–3]. In this context, accelerating the adjustment of energy structures and promoting the clean, low-carbon, and high-efficiency transformation of energy systems has become a dominant



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trend in global energy development [4]. Renewable energy sources represented by wind and solar power have achieved rapid development due to their abundant reserves, environmental benignity, and sustainable characteristics, gradually emerging as core components of future energy systems. However, compared with conventional dispatchable power sources, wind and solar power generation are highly dependent on meteorological conditions, diurnal variations, and seasonal fluctuations. Their inherent volatility, intermittency, and stochasticity lead to a poor real-time matching capability between power output and grid load demand.

Energy storage technologies enable temporal and spatial regulation and redistribution of electric power via reversible energy charging and discharging behaviors, which plays a vital role in peak shaving and valley filling, mitigating output fluctuations of renewable energy, improving power system flexibility, and guaranteeing the safe and stable operation of power grids [5,6]. Among various electrochemical energy storage technologies, flow batteries exhibit prominent advantages including decoupled power and capacity design, superior safety, long cycle lifespan, and excellent environmental adaptability, rendering them promising candidates for large-scale long-duration energy storage scenarios [7,8]. Flow batteries are classified into non-aqueous and aqueous systems according to electrolyte properties, which differ substantially in electrochemical window, energy density, safety level, capital cost, and applicable operating conditions [9]. Benefiting from high electrolyte conductivity, intrinsic operational safety, low cost, and mild operating conditions, aqueous flow batteries have attracted extensive research attention for large-scale energy storage applications [10–13].

Among diverse emerging aqueous flow battery configurations, neutral iron-sulfur flow batteries have gradually demonstrated remarkable research significance and application potential in recent years, attributed to the wide availability of active species, low capital expenditure, environmental benignity, and mild operating environments [14]. As illustrated in Figure 1, ferricyanide/ferrocyanide redox couples generally serve as the positive active species, while sulfide/polysulfide redox couples function as the negative active species within this system [15]. The ferricyanide/ferrocyanide redox pair possesses favorable chemical stability and cycling reversibility under neutral or near-neutral aqueous conditions [16–19]. Meanwhile, polysulfides feature abundant raw material reserves, low material cost, and high theoretical specific capacity. The favorable potential matching between these two redox pairs offers a feasible strategy for constructing aqueous flow battery systems with superior safety, low corrosivity, and scalable manufacturing capacity. Recently, electrode modification strategies have been widely adopted to enhance the interfacial reactivity and electrochemical performance of flow batteries [20]. In order to address the persistent issues of severe shuttle effect and sluggish interfacial reaction kinetics that polysulfides still encounter during practical operation, previous researchers have conducted extensive studies. However, currently, sulfide-modified materials such as WS_2 [21] and NiMoS [22] still involve complex preparation processes and high costs, and their applicability and regulatory mechanisms in the interfacial reactions of polysulfides in neutral iron-sulfur flow batteries remain rather limited. In contrast, nickel sulfide materials feature advantages such as lower raw material costs, relatively simple preparation methods, and good electrical conductivity [23,24]. Nevertheless, the limited solubility of ferricyanide/ferrocyanide species on the positive side (typically below 0.5 M), which restricts the enhancement of specific capacity and energy density, acts as a critical bottleneck hindering the performance optimization of this system.

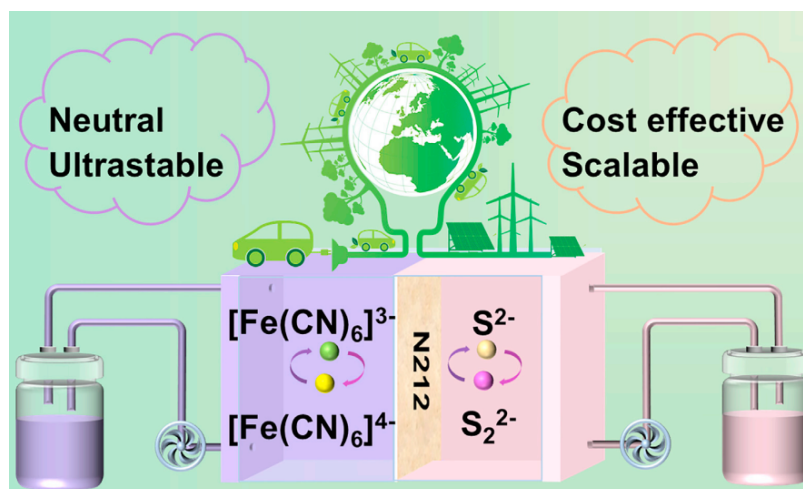


Figure 1. Schematic diagram of the neutral polysulfide/ferricyanide flow battery system. Reprinted with permission from Ref. [15]. Copyright 2021, The Authors.

Redox-targeted reactions provide a novel research insight to address the aforementioned challenges [25]. Different from conventional flow batteries, redox-targeted flow batteries break the conventional energy storage mode that relies merely on soluble active species by introducing solid energy storage materials into the electrolyte reservoir, thereby achieving capacity enhancement through chemical redox reactions between soluble redox mediators and solid active materials. As illustrated in Figure 2, soluble mediators first exchange electrons at the electrode, then undergo chemical redox reactions with solid materials in the reservoir, completing a cyclic electron-transfer loop.

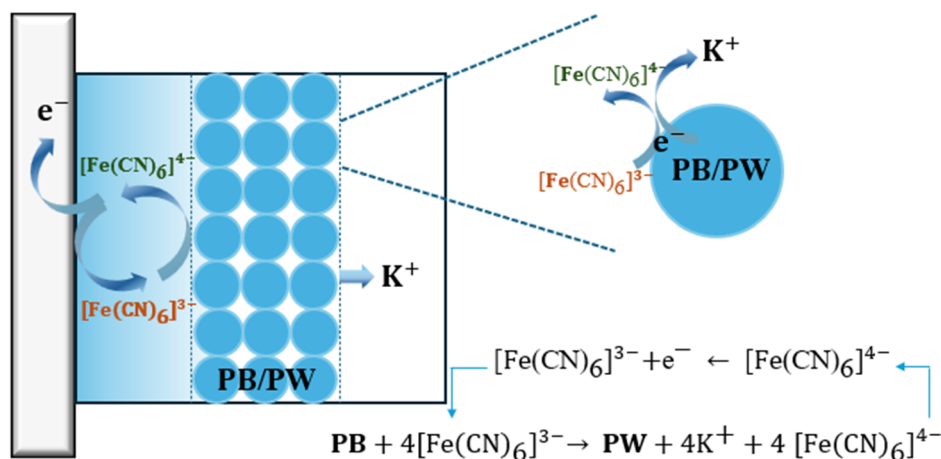


Figure 2. Schematic illustration of the redox-targeted reaction mechanism.

In 2017, Fan et al. pioneered the incorporation of Prussian blue into redox-targeted flow battery configurations for the construction of low-cost, high-energy-density flow batteries and solar-rechargeable flow batteries, which validates the promising application prospect of Prussian blue analogues in this research field [26,27]. In 2019, Chen et al. [28] reported a stable and fast unimolecular redox-targeted flow battery based on the $[\text{Fe}(\text{CN})_6]^{4-/3-}$ couple and Prussian blue within a low-cost neutral aqueous system. The as-fabricated battery delivered a volumetric capacity of 61.6 Ah L^{-1} with a capacity retention rate exceeding 99.99% over cycling. This landmark work directly verifies the favorable potential matching and rapid solid-liquid redox kinetics between the ferricyanide/ferricyanide pair and Prussian blue, thereby offering solid theoretical support for the utilization of Prussian blue as a capacity booster on the ferricyanide positive side in the present study. Collectively, the aforementioned investigations demonstrate that redox-targeted reactions possess remarkable application potential in neutral aqueous flow batteries [29–34]. Nevertheless, prominent limitations remain in existing investigations. Although Fan et al. constructed Prussian blue-based redox-targeted flow batteries, their work lacks systematic optimization of solid energy-storage material loading fractions. Chen et al. mainly verified the feasibility of redox-targeted reactions between $[\text{Fe}(\text{CN})_6]^{4-/3-}$ and Prussian blue, yet did not systematically elucidate how Prussian blue dosage governs reaction kinetics, capacity utilization efficiency and cycling stability. From a practical application perspective, insufficient Prussian blue loading yields marginal capacity improvement, while excessive loading may induce inadequate solid-liquid contact and severe agglomeration of solid energy storage materials. Accordingly, revealing the quantitative correlation between Prussian blue content and targeted reaction efficiency, positive-side capacity as well as overall cell performance is of great significance for guiding the engineering design of redox-targeted flow batteries, which also constitutes a critical research gap urgently requiring in-depth exploration in this field [35].

This work targets neutral iron-sulfur flow batteries, focusing on the capacity enhancement mechanism and dosage optimization of Prussian blue on the positive electrode side. Furthermore, the optimized Prussian blue is integrated into the full battery system for comprehensive performance evaluation. The main research contents are summarized as follows:

(1) Investigation on redox-targeted reaction behavior and dosage optimization of Prussian blue

Aiming at the limited positive capacity of ferricyanide caused by the solubility constraint of liquid-phase active species, a redox-targeted symmetric flow battery model system is constructed with the $[\text{Fe}(\text{CN})_6]^{4-}/[\text{Fe}(\text{CN})_6]^{3-}$ couple serving as liquid redox mediators and Prussian blue as solid energy storage material. The activation behavior of Prussian blue during cycling and its structural evolution toward Prussian white are systematically explored. On this basis, the effects of ferricyanide concentration and Prussian blue loading

fraction on capacity delivery performance are further evaluated, laying a theoretical foundation for developing capacity enhancement strategies for the positive electrode compartment.

(2) Performance Study of Neutral Iron-Sulfur Flow Batteries Based on Optimized Prussian Blue Loading

On the basis of the above research, Prussian blue with the optimal loading is introduced into the neutral iron-sulfur flow battery system, where potassium sulfide serves as the negative electrolyte, to verify the full-cell electrochemical performance. The negative electrolyte is potassium sulfide solution with a measured pH of 7–8. The system is defined as a neutral iron-sulfur flow battery because it operates in a near-neutral pH range, different from conventional strongly alkaline iron-sulfur flow batteries. Polarization curves, power density curves, voltage-capacity profiles and cycling efficiencies are comprehensively analyzed. Furthermore, the variation laws of full-cell energy efficiency under different current densities and temperatures are systematically explored to evaluate the comprehensive performance response of the battery system under diverse operating conditions.

2. Experimental

2.1. Experimental Materials and Reagents

The primary raw materials and chemical reagents adopted in this work are summarized in Table 1.

Table 1. Information of experimental materials.

Material Name	Specification	Manufacturer
Nickel chloride hexahydrate	Analytical pure	Aladdin Reagents Co., Ltd., Shanghai, China.
Thiourea	Analytical pure	Macklin Biochemical Co., Ltd., Shanghai, China.
Anhydrous ethanol	Analytical pure	Sinopharm Chemical Reagent Co., Ltd., Beijing, China.
Deionized water	Grade I	Resjete Water Purification Technology Co., Ltd., Xiamen, China.
Potassium chloride	Analytical pure	Sinopharm Chemical Reagent Co., Ltd., Beijing, China.
Potassium ferrocyanide	99% purity	Macklin Biochemical Co., Ltd., Shanghai, China.
Potassium ferricyanide	Analytical pure	Aladdin Reagents Co., Ltd., Shanghai, China.
Anhydrous potassium sulfide	Analytical pure	Macklin Biochemical Co., Ltd., Shanghai, China.
Graphite felt	GFA 6 EA, 2.5 mm	SGL Carbon SE, Wiesbaden, Germany.
Nafion membrane	Nafion 212	DuPont, Wilmington, USA.

2.2. Fabrication of NiS_x -GF Composite Electrodes

The NiS_x -GF composite electrodes were fabricated in situ via a hydrothermal route. Briefly, graphite felts were cut into square pieces with dimensions of $2\text{ cm} \times 2\text{ cm}$, followed by sequential rinsing with anhydrous ethanol and deionized water to eliminate surface impurities and organic contaminants. The cleaned graphite felts were then dried for subsequent use. Subsequently, 118.80 mg of nickel chloride hexahydrate (nickel precursor) and 152.20 mg of thiourea (sulfur precursor) were dissolved in 40 mL deionized water under magnetic stirring to obtain a homogeneous precursor solution. The preprocessed graphite felts were fully immersed in the as-prepared solution and transferred into a Teflon-lined autoclave for a hydrothermal reaction at $160\text{ }^\circ\text{C}$ over 6 h.

Upon the completion of the hydrothermal reaction, the autoclave was naturally cooled down to ambient temperature. The as-obtained samples were taken out and repeatedly rinsed with deionized water to eliminate unreacted residual species on the surface, followed by a drying procedure. The modified graphite felts were further cut into $1\text{ cm} \times 1\text{ cm}$ slices for subsequent electrochemical measurements, yielding nickel sulfide-modified graphite felt composite electrodes denoted as NiS_x -GF.

During the hydrothermal treatment, thiourea acts as a slow-releasing sulfur source that gradually decomposes to generate S^{2-} ions, which facilitates the in-situ sulfurization of Ni^{2+} on the surface of graphite felt fibers. Owing to the influences of reaction kinetics, local chemical environment and interfacial nucleation-growth behaviors during sulfurization, the resultant products may simultaneously contain Ni_3S_2 , NiS and minor low-crystallinity or amorphous sulfide layers. Accordingly, such non-stoichiometric nickel sulfides are collectively referred to as NiS_x throughout this work.

2.3. Preparation of Electrolyte for Symmetric Flow Batteries and Investigation of Prussian Blue (PB)-Added Systems

2.3.1. Assembly of Symmetric Flow Batteries and Preparation of Electrolytes

In this work, the $[\text{Fe}(\text{CN})_6]^{4-}/[\text{Fe}(\text{CN})_6]^{3-}$ redox couple was selected as the liquid-phase redox mediator, and PB served as the solid energy storage material to construct redox-targeted symmetric flow batteries. Potassium

chloride (KCl) was adopted as the supporting electrolyte to elevate the ionic conductivity and stabilize the ionic strength of the electrolyte solution.

To explore the effect of ferricyanide concentration on the activation behavior of PB, electrolytes with varied concentrations of potassium ferrocyanide and potassium ferricyanide were prepared separately. Specifically, a certain mass of potassium ferrocyanide was dissolved in deionized water containing 1 M KCl supporting electrolyte to yield ferrocyanide electrolytes at concentrations of 0.1 M, 0.2 M and 0.3 M. Potassium ferricyanide electrolytes were fabricated following an identical protocol.

A symmetric flow battery was first assembled to investigate the practical activation performance of PB in the $[\text{Fe}(\text{CN})_6]^{4-}/[\text{Fe}(\text{CN})_6]^{3-}$ liquid mediator. Identical $\text{NiS}_x\text{-GF}$ composite electrodes were deployed on both sides of the symmetric cell, while the electrolyte volumes of the two compartments were differentiated, with the smaller-volume compartment acting as the capacity-limited side. The detailed electrolyte configuration was as follows: the positive compartment contained 10 mL potassium ferrocyanide solution with 1 M KCl, and the negative compartment contained 20 mL potassium ferricyanide solution with 1 M KCl.

For symmetric cell assembly, $\text{NiS}_x\text{-GF}$ composite electrodes and Nafion 212 membrane were utilized as the electrode and separator, respectively. Galvanostatic charge-discharge measurements were implemented at a constant current of 40 mA, with cut-off voltages set at 0.4 V (charge) and −0.4 V (discharge). After the cell completed the first cycle, a predetermined amount of PB was introduced into the positive electrolyte reservoir prior to the start of the second cycle to further characterize PB activation. By comparing the charge-discharge duration and capacity variation before and after PB addition, it was determined whether PB could be effectively activated by the $[\text{Fe}(\text{CN})_6]^{4-}/[\text{Fe}(\text{CN})_6]^{3-}$ mediator and participate in redox-targeted energy storage.

2.3.2. Addition Scheme of Prussian Blue

PB powder was directly added into the positive electrolyte reservoir and served as the solid energy storage material for the positive compartment. To investigate the influence of PB loading on the capacity enhancement via redox-targeted reactions, PB was introduced at molar ratios of 10%, 20%, 30% and 40% relative to potassium ferrocyanide.

During electrochemical tests, PB remained stationary as solid sediment in the reservoir without circulating along with the electrolyte. Redox-targeted reactions proceeded between the solid PB and soluble $[\text{Fe}(\text{CN})_6]^{4-}/[\text{Fe}(\text{CN})_6]^{3-}$ mediators, enabling electron transfer and delivering extra specific capacity.

2.4. Full-Cell Assembly and Electrolyte Preparation

After determining the representative working concentration and optimal loading of PB, PB with the optimized dosage was incorporated into the positive electrolyte reservoir of the neutral iron-sulfur flow battery, and $\text{NiS}_x\text{-GF}$ composite electrodes were employed as negative electrodes for full-cell performance validation.

The positive electrolyte of the neutral iron-sulfur flow battery consisted of 10 mL 0.3 M potassium ferrocyanide solution containing 1 M KCl supporting electrolyte, while the negative electrolyte was 20 mL 0.5 M potassium sulfide solution with 1 M KCl. Nafion 212 membrane was utilized as the separator.

3. Results and Discussion

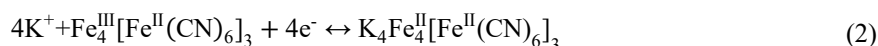
3.1. Fundamental Analysis of Potential Matching and Reaction Kinetics for Prussian Blue–Ferricyanide Systems

Cyclic voltammetry (CV) measurements were first carried out on the soluble $[\text{Fe}(\text{CN})_6]^{4-}/[\text{Fe}(\text{CN})_6]^{3-}$ mediator and solid PB to verify whether PB can engage in energy storage via redox-targeted reactions within the ferricyanide system, as illustrated in Figure 3a. A distinct pair of redox peaks can be observed for the $[\text{Fe}(\text{CN})_6]^{4-}/[\text{Fe}(\text{CN})_6]^{3-}$ couple over the tested potential window, demonstrating favorable electrochemical reversibility of this liquid redox mediator. Clear redox signals were also detected for PB, whose peak potentials closely overlap with those of the ferricyanide redox pair. This observation confirms satisfactory potential matching between the aqueous $[\text{Fe}(\text{CN})_6]^{4-}/[\text{Fe}(\text{CN})_6]^{3-}$ mediator and the PB/PW redox couple, which thermodynamically validates the feasibility of PB participating in redox-targeted energy storage reactions.

The redox reaction of the liquid-phase $[\text{Fe}(\text{CN})_6]^{4-}/[\text{Fe}(\text{CN})_6]^{3-}$ mediator can be expressed as follows:



The reversible conversion between PB and PW can be expressed as follows:



The two reactions above are coupled through electron transfer between the liquid mediator and solid PB, forming a complete redox-targeting cycle. Taking discharge as an example: the reduced mediator $[\text{Fe}(\text{CN})_6]^{4-}$, generated at the electrode, diffuses to the electrolyte reservoir and donates electrons to solid PB, reducing it to Prussian white (PW), while itself being re-oxidized to $[\text{Fe}(\text{CN})_6]^{3-}$. During charge, the reverse occurs: the oxidized mediator $[\text{Fe}(\text{CN})_6]^{3-}$ extracts electrons from PW, regenerating PB and being reduced back to $[\text{Fe}(\text{CN})_6]^{4-}$. Thus, electrons are shuttled between the electrode and the solid material via the liquid mediator, enabling PB to participate in energy storage without direct electrode contact, thereby overcoming the capacity limitation imposed by the solubility of the liquid-phase active species. To further investigate the electrochemical kinetic characteristics of the $[\text{Fe}(\text{CN})_6]^{4-}/[\text{Fe}(\text{CN})_6]^{3-}$ system, cyclic voltammetry curves at various scan rates were recorded. The peak current densities were plotted against the square root of scan rate ($v^{1/2}$), as displayed in Figure 3b. Both anodic and cathodic peak current densities exhibit satisfactory linear correlations with $v^{1/2}$, revealing that the electrode process of this redox couple is predominantly diffusion-controlled. As the effective electrode area used in this work was 1 cm^2 , this relationship can be described by the Randles-Sevcik equation:

$$i_p = 2.69 \times 10^5 A n^{3/2} c D^{1/2} v^{1/2} \quad (3)$$

where

- $i_p(\text{A})$ —peak current;
- $A(\text{cm}^2)$ —electrode area;
- $c(\text{mol} \cdot \text{cm}^{-3})$ —concentration of active species;
- $D(\text{cm}^2 \cdot \text{s}^{-1})$ —diffusion coefficient;
- $v(\text{V} \cdot \text{s}^{-1})$ —scan rate;
- n —number of electrons transferred in the redox reaction.

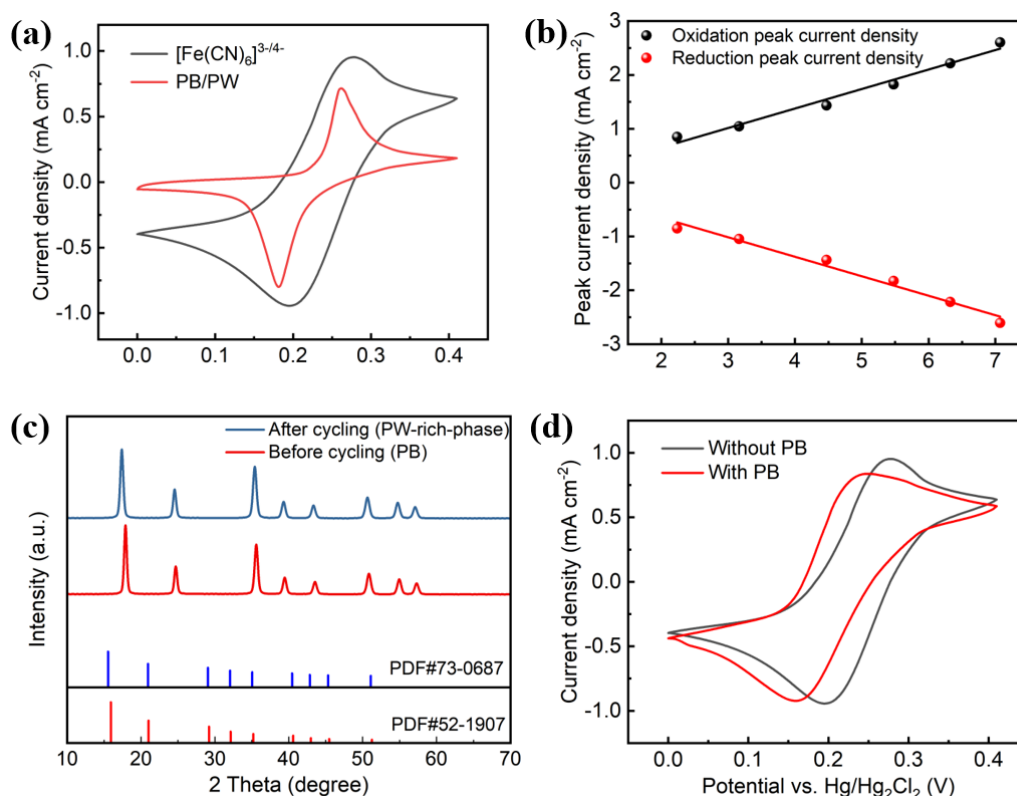


Figure 3. (a) Cyclic voltammetry curves of ferricyanide system and Prussian blue; (b) Linear relationships between anodic/cathodic peak current densities and the square root of scan rate for the ferricyanide system; (c) XRD patterns of solid Prussian blue samples before and after cycling; (d) Cyclic voltammetry curves of the $[\text{Fe}(\text{CN})_6]^{4-}/[\text{Fe}(\text{CN})_6]^{3-}$ system with and without Prussian blue addition.

The above results confirm that the mediator's electron transfer capacity is concentration-dependent: low concentrations limit PB activation, whereas higher concentrations enable effective PB participation, providing the kinetic basis for subsequent concentration-dependent studies.

3.2. Structural Transformation and Electrochemical Evidence of Redox-Targeted Reactions for Prussian Blue

XRD characterizations were performed on solid PB samples before and after electrochemical cycling to confirm that PB can take part in energy storage via redox-targeted reactions in the $[\text{Fe}(\text{CN})_6]^{4-}/[\text{Fe}(\text{CN})_6]^{3-}$ liquid mediator, as shown in Figure 3c. The dominant diffraction peaks of the pristine sample well match the standard pattern of Prussian blue (PDF#52-1907), verifying that the as-introduced solid particles are pure crystalline PB phase. After cycling, all major diffraction peaks shift toward lower angles. The observed peak shift is attributable to lattice expansion resulting from the reduction of Fe(III) to Fe(II) and the concomitant intercalation of K^+ ions into the PB framework, which increases the unit cell parameter and consequently shifts diffraction peaks to lower 2θ angles. This structural evidence confirms that PB undergoes reversible phase transition driven by the mediator rather than remaining inert. According to the energy storage mechanism of the PB/PW redox pair, soluble $[\text{Fe}(\text{CN})_6]^{4-}$ acts as a reduced electron shuttle during discharge and exchanges electrons with solid PB in the positive reservoir, gradually reducing PB to PW. Upon charging, oxidized $[\text{Fe}(\text{CN})_6]^{3-}$ serves as the electron carrier and re-oxidizes PW back to PB. Therefore, the PW enrichment detected in post-cycling solid samples is consistent with the reversible PB/PW transformation enabled by redox-targeted reactions, offering direct structural evidence that PB participates in energy storage.

Comparative cyclic voltammetry measurements of the $[\text{Fe}(\text{CN})_6]^{4-}/[\text{Fe}(\text{CN})_6]^{3-}$ system with and without PB were conducted to further electrochemically verify the participation of PB, as displayed in Figure 3d. Distinct variations are observed in the CV profile after PB addition relative to the PB-free system, where both anodic and cathodic peaks shift negatively, demonstrating that PB participates in the redox equilibrium, with the solid-phase PB/PW couple establishing a competing electrochemical pathway that modifies the apparent equilibrium potential of the soluble $[\text{Fe}(\text{CN})_6]^{4-}/[\text{Fe}(\text{CN})_6]^{3-}$ mediator. This phenomenon confirms that PB is not an inert solid within the system but can exchange electrons with the liquid redox mediator, thereby shifting the equilibrium potential of the $[\text{Fe}(\text{CN})_6]^{4-}/[\text{Fe}(\text{CN})_6]^{3-}$ couple. Such electrochemical behavior agrees well with the structural evidence of PB-to-PW conversion obtained from XRD analysis. The consistent results jointly confirm that PB can engage in redox-targeted energy storage mediated by aqueous $[\text{Fe}(\text{CN})_6]^{4-}/[\text{Fe}(\text{CN})_6]^{3-}$.

3.3. Effect of Ferrocyanide Concentration on the Activation Behavior of Prussian Blue

After verifying the favorable potential matching between PB and the $[\text{Fe}(\text{CN})_6]^{4-}/[\text{Fe}(\text{CN})_6]^{3-}$ couple, symmetric flow batteries were assembled with identical electrolyte concentrations at both positive and negative sides to explore how the concentration of liquid redox mediator influences the practical activation of PB. Figure 4a–c present the galvanostatic charge-discharge profiles of symmetric cells before and after PB addition at various ferrocyanide concentrations. The results reveal that negligible variations in charge and discharge durations are observed upon PB introduction for the 0.1 M and 0.2 M ferrocyanide systems. In contrast, both charging and discharging durations are remarkably prolonged with PB added to the 0.3 M electrolyte, demonstrating that PB can be efficiently activated and contribute to energy storage at this mediator concentration.

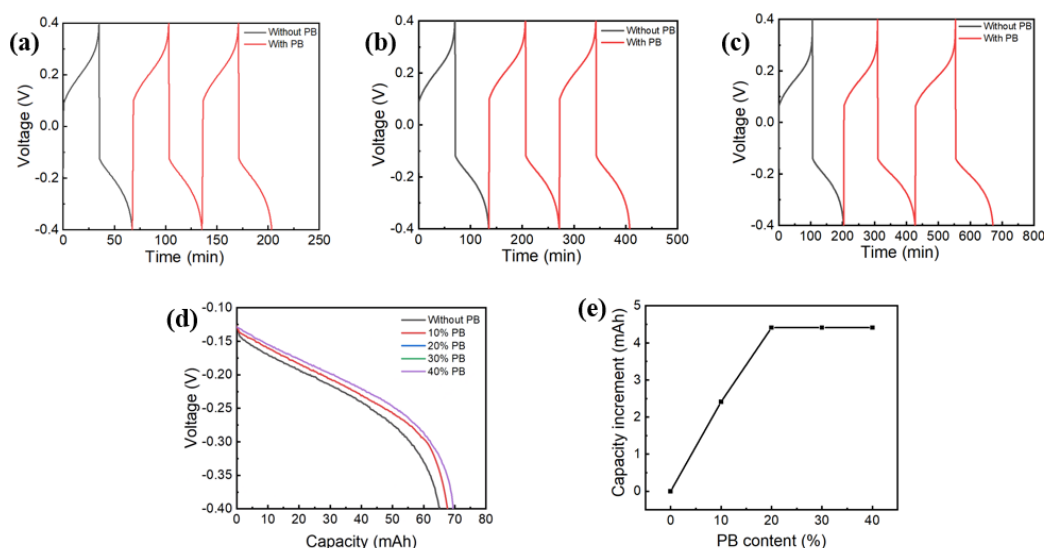


Figure 4. Galvanostatic charge-discharge profiles of symmetric flow batteries at ferrocyanide concentrations of 0.1 M (a), 0.2 M (b) and 0.3 M (c); (d) Voltage-capacity curves of symmetric flow batteries with various PB dosages; (e) Capacity increment variations of symmetric flow batteries under different PB addition amounts.

The above results reveal that the concentration of liquid redox mediator exerts a pronounced influence on the practical activation of PB under the fixed current, electrolyte volume and testing duration adopted in this work. At ferrocyanide concentrations of 0.1 M and 0.2 M, although redox-targeted reactions are thermodynamically feasible, the electron exchange capacity of the liquid mediator remains insufficient to drive obvious capacity contribution from PB within the experimental timescale. In contrast, elevating the mediator concentration to 0.3 M enhances the electron transfer kinetics of the liquid phase, enabling faster and more complete activation of PB, which is manifested as substantially prolonged charge/discharge durations and increased total capacity.

It should be emphasized that the concentration gradient was designed to compare representative low, medium and high concentrations, identify whether PB can be effectively activated in the $[\text{Fe}(\text{CN})_6]^{4-}/[\text{Fe}(\text{CN})_6]^{3-}$ system, and screen a working concentration for subsequent optimization of PB dosage, rather than precisely defining the critical concentration threshold for remarkable PB activation. Therefore, three progressive concentrations (0.1 M, 0.2 M and 0.3 M) were selected to investigate the overall trend of PB activation behavior with rising mediator concentration. Experimental data show that distinct capacity contributions from PB are only observed at 0.3 M within the tested concentration range of 0.1–0.3 M; accordingly, 0.3 M was chosen as the representative working concentration for the subsequent PB dosage optimization study. Although finer concentration gradients would help narrow down the exact concentration range for prominent PB activation, they do not alter the core conclusion of this work that higher mediator concentrations facilitate efficient PB activation.

In summary, ferrocyanide concentration is a vital parameter governing PB activation performance. Within the concentration window examined herein, the 0.3 M aqueous $[\text{Fe}(\text{CN})_6]^{4-}/[\text{Fe}(\text{CN})_6]^{3-}$ mediator enables significant capacity output from PB and serves as a suitable working concentration for further PB dosage optimization. This finding is consistent with the kinetic characteristics derived from preceding cyclic voltammetry analysis, demonstrating that liquid mediator concentration not only governs the electrochemical response intensity of the system but also directly determines the extent of capacity release from PB in symmetric flow batteries.

3.4. Effect of Prussian Blue Dosage on Capacity Release

Having confirmed the efficient activation of PB in the 0.3 M ferrocyanide system, the PB dosage was determined on the basis of the molar quantity of potassium ferrocyanide, yielding loadings of 10% (257.80 mg), 20% (515.50 mg), 30% (773.30 mg), and 40% (1031.10 mg). The influence of PB loading on the capacity release behavior was then further examined. Figure 4d displays the voltage-capacity curves of symmetric flow batteries with PB molar ratios of 10%, 20%, 30% and 40%, and Figure 4e illustrates the corresponding variation of capacity increment.

Under galvanostatic charge–discharge operation, the discharge capacity is defined as:

$$Q = \frac{It}{3600} \quad (4)$$

where $Q(\text{mAh})$ —discharge capacity;

$I(\text{mA})$ —current;

$t(\text{s})$ —discharge time.

To quantitatively compare the capacity enhancement at various PB dosages, the capacity increment induced by PB is defined as:

$$\Delta Q_{\text{PB}} = Q_{\text{with PB}} - Q_{\text{blank}} \quad (5)$$

where $Q_{\text{with PB}}(\text{mAh})$ —discharge capacity after PB addition;

$Q_{\text{blank}}(\text{mAh})$ —baseline discharge capacity of the PB-free cell.

The relative capacity increment ratio is further formulated as:

$$\eta_{\text{inc}} = \frac{\Delta Q_{\text{PB}}}{Q_{\text{blank}}} \times 100\% \quad (6)$$

Experimental results reveal that the discharge capacity rises remarkably when the PB molar ratio increases from 10% to 20%, indicating that more PB particles participate in redox-targeted reactions and deliver extra capacity. The capacity growth slows down as the PB loading is further raised from 20% to 30%, and the capacity at 40% PB is nearly identical to that at 30%. This trend demonstrates that the capacity contribution plateaus gradually with increasing PB dosage rather than rising indefinitely, which implies an effective utilization window and an upper capacity limit for PB.

This saturation behavior can first be interpreted by the limited electron reservoir of the liquid mediator. The capacity-limited compartment of the symmetric cell contains 10 mL of 0.3 M ferrocyanide electrolyte, and the total amount of available liquid mediator is calculated as:

$$n_{\text{med}} = cV = 0.3 \times 0.010 = 0.003 \text{ mol} \quad (7)$$

Since the $[\text{Fe}(\text{CN})_6]^{4-}/[\text{Fe}(\text{CN})_6]^{3-}$ redox couple undergoes a one-electron transfer reaction, the maximum available electron reservoir from the capacity-limited side is:

$$n_{\text{e, med}} = 0.003 \text{ mol e}^- \quad (8)$$

In contrast, the reversible conversion between PB and PW involves a four-electron redox process, so the theoretical electron consumption for complete PB conversion is expressed as:

$$n_{\text{e, PB}} = 4n_{\text{PB}} \quad (9)$$

Calculations show that the theoretical electron demand of PB accounts for 40%, 80%, 120% and 160% of the mediator's electron reservoir at 10–40% PB loadings, respectively. Thus, at 30% and 40%, the electron requirement exceeds the available supply, leaving excess PB unactivated.

Second, the capacity plateau is closely associated with mass transfer limitations at the solid–liquid interface. Elevated PB loading aggravates particle sedimentation and agglomeration inside the electrolyte reservoir. Outer PB granules preferentially contact the electrolyte and undergo redox reactions, while inner particles suffer insufficient liquid contact and cannot be fully utilized. As a result, the effective reactive surface area stops increasing linearly with the total PB mass. In other words, further adding PB from 20% to 30% and 40% mainly introduces inactive solid reserves that barely contribute to accessible discharge capacity.

The data in Figure 4d,e reveal that the discharge capacity increases markedly when the PB ratio rises from 10% to 20%, yet exhibits negligible further increment beyond 20%. This plateau indicates that 20% represents the upper bound of effective PB utilization under the present conditions. Consequently, 20% is identified as the optimal loading.

In summary, this section verifies that PB can participate in energy storage via redox-targeted reactions mediated by aqueous $[\text{Fe}(\text{CN})_6]^{4-}/[\text{Fe}(\text{CN})_6]^{3-}$. The quantitative relationships between mediator concentration/PB dosage and PB activation as well as capacity contribution are systematically clarified, laying an experimental foundation for the capacity-boosting strategy by introducing PB into full flow batteries.

3.5. Synergistic Effect and Operating Condition Adaptability of the Integrated System

3.5.1. Electrochemical Performance of Full Cells Incorporating Optimized PB Loading

Within the scope of the present investigation, the 0.3 M ferrocyanide electrolyte was selected as the representative working concentration for optimizing PB loading. Notably, the cell capacity rises substantially when the PB molar ratio increases from 10% to 20%, while further elevating the ratio to 30% and 40% yields negligible additional capacity gain, demonstrating that the system approaches the upper limit of effective PB utilization at 20%. Accordingly, a 20% PB molar ratio was adopted as the optimal addition protocol for subsequent full-cell measurements.

On the basis of the above findings, the optimal PB loading strategy was identified as the key to enhancing the cathodic capacity. As the critical component of the targeting mechanism, its influence on the full-cell operation was then further investigated. In practical implementation, PB was supplemented at a molar ratio of 20% relative to potassium ferrocyanide in the positive electrolyte. For 10 mL of 0.3 M potassium ferrocyanide catholyte, the corresponding PB dosage was 515.50 mg. Solid PB particles were loaded into the positive electrolyte reservoir instead of circulating through the cell stack. Electron exchange occurred between stationary PB and soluble $[\text{Fe}(\text{CN})_6]^{4-}/[\text{Fe}(\text{CN})_6]^{3-}$ mediators to realize redox-targeted energy storage. Unless otherwise specified, this PB addition strategy was applied to all subsequent full-cell tests with PB-based capacity enhancement, ensuring consistent and comparable experimental data across different test conditions.

Polarization curves and power density profiles of full cells with and without optimal PB loading were characterized to systematically evaluate the effect of PB incorporation on the performance of neutral iron-sulfur flow batteries, as illustrated in Figure 5a. The two cells exhibit slightly different initial open-circuit voltages (OCVs). As current density increases, however, the PB-added cell displays milder voltage decay and a broader effective operating window, revealing superior polarization behavior and power output under moderate and high current densities. Although the cyclic voltammetry test in Figure 3d confirms a negative shift in the equilibrium potential of the $[\text{Fe}(\text{CN})_6]^{4-}/[\text{Fe}(\text{CN})_6]^{3-}$ couple after PB addition, this potential variation barely alters the full-cell OCV, yet substantially improves capacity utilization and discharge performance during cycling.

According to the power density plots, the baseline cell without PB delivers a peak power density of approximately $12.00 \text{ mW} \cdot \text{cm}^{-2}$, whereas the cell integrated with optimized PB achieves an elevated peak power density of $14.58 \text{ mW} \cdot \text{cm}^{-2}$. This corresponds to a 21.5% improvement in peak power density, which is primarily

attributable to the enhanced positive-side capacity and reduced polarization losses resulting from PB incorporation. This comparison verifies that incorporating appropriate PB on the positive side greatly boosts the power capability of cells equipped with NiS_x-GF composite negative electrodes. Furthermore, the PB-modified cell sustains stable discharge at higher current densities, which corresponds to an improved limiting operating current density. Collectively, these results confirm that the introduction of PB at the positive compartment not only maximizes the peak power output of full cells but also facilitates stable operation under high-rate conditions.

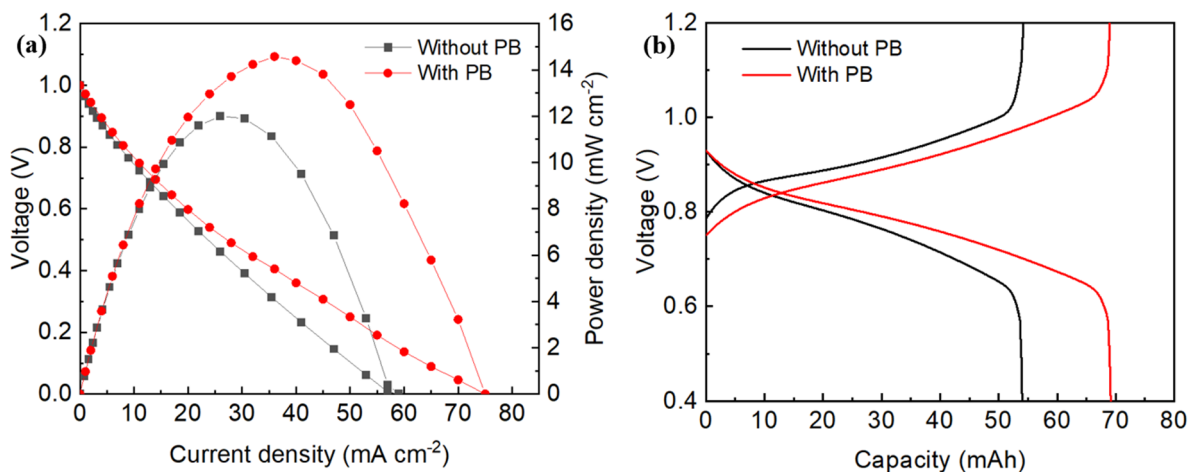


Figure 5. (a) Polarization curves and power density profiles of neutral iron-sulfur flow batteries with and without Prussian blue; (b) Voltage-capacity curves of neutral iron-sulfur flow batteries with and without Prussian blue.

Voltage-capacity measurements were carried out to further compare the charge-discharge behaviors of full cells with and without PB, as displayed in Figure 5b. The discharge capacity is significantly enhanced after PB incorporation, with the maximum capacity rising from 54.40 mAh (PB-free cell) to 69.66 mAh. This proves that PB effectively expands the accessible capacity of the positive compartment and delivers higher overall capacity for the full cell. Meanwhile, the cell loaded with PB exhibits a reduced charging plateau while the discharge plateau remains nearly unchanged, which indicates that PB mainly optimizes the voltage profile during charging and alleviates cell polarization to a certain extent. This observation coincides with the foregoing polarization and power density test results, demonstrating that the introduction of optimal PB dosage not only promotes capacity release on the positive side but also improves the charging voltage response of the full cell.

The efficiency evolution during long-term cycling was further analyzed and presented in Figure 6. As illustrated in Figure 6a, both cells maintain high Coulombic efficiency with minor fluctuations over cycles, which reflects favorable charge utilization and cycling stability for both systems. Nevertheless, the PB-integrated cell delivers slightly higher average Coulombic efficiency than the blank cell. After PB participates in positive-side energy storage via redox-targeted reactions, the utilization of iron-based active species is improved, and the charge-discharge mismatch induced by limited positive capacity is mitigated, thereby sustaining stable and superior charge utilization throughout cycling.

Figure 6b compares the voltage efficiency variations of neutral iron-sulfur flow batteries with and without PB. The PB-added cell retains consistently higher voltage efficiency across all cycles, with a more remarkable improvement than that of Coulombic efficiency. This reveals that PB incorporation reduces voltage loss and suppresses polarization during charge and discharge. Consistent with the lowered charging plateau and elevated capacity output observed in Figure 5b, these results confirm that optimized PB loading boosts positive capacity utilization and optimizes the charging voltage performance of the full cell simultaneously.

Figure 6c presents the energy efficiency variations of neutral iron-sulfur flow batteries with and without PB. The cell loaded with PB maintains persistently higher energy efficiency throughout cycling with a milder degradation trend. Since energy efficiency is jointly governed by Coulombic efficiency and voltage efficiency, combined with the data in Figure 6a,b, the prominent elevation in energy efficiency after PB addition primarily stems from the remarkable improvement in voltage efficiency, accompanied by a slight enhancement in Coulombic efficiency. In other words, while boosting the deliverable capacity of the positive compartment, PB incorporation does not introduce severe extra polarization. Instead, it optimizes the energy storage reaction on the positive side and the matching degree of overall cell redox reactions, thereby elevating the comprehensive energy utilization efficiency.

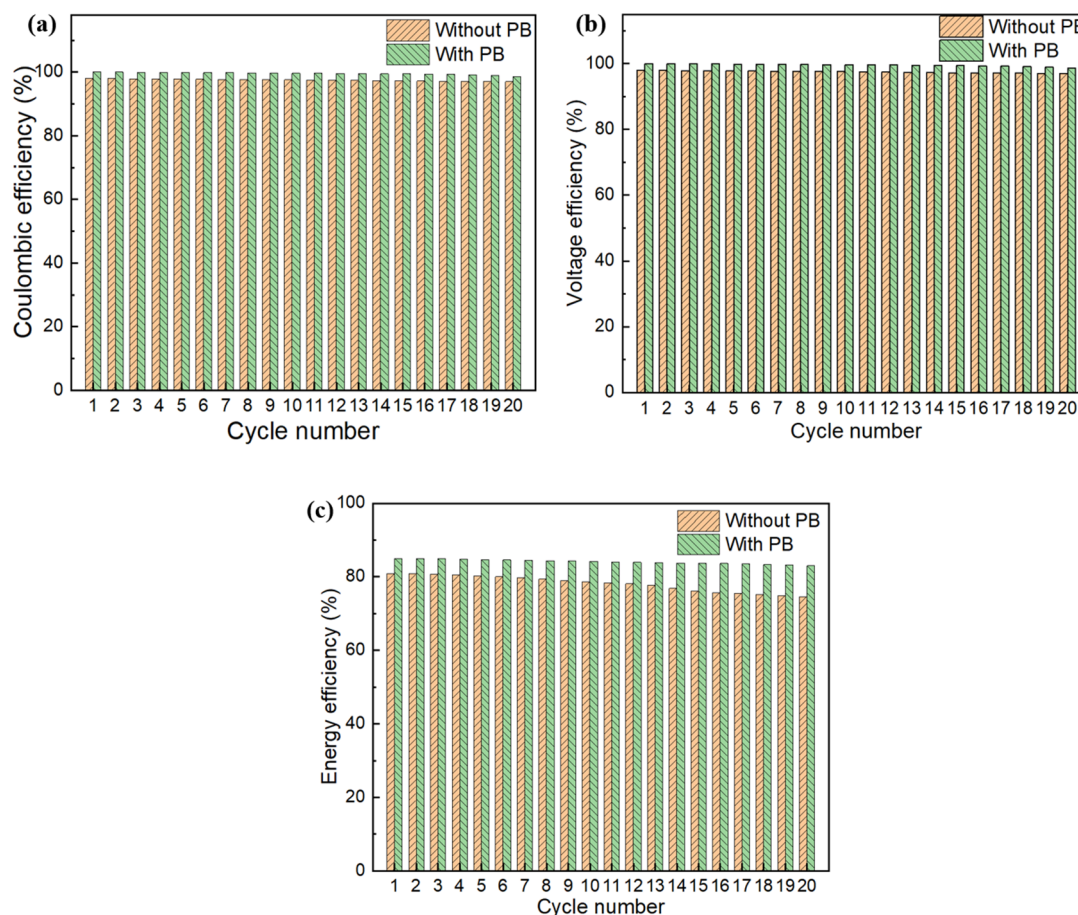


Figure 6. (a) Coulombic efficiency, (b) voltage efficiency and (c) energy efficiency evolutions of neutral iron-sulfur flow batteries with and without Prussian blue.

In summary, introducing PB with optimized dosage simultaneously enhances the capacity output, polarization performance and efficiency retention of neutral iron-sulfur flow batteries. The enlarged discharge capacity and improved voltage efficiency constitute the dominant contributors to higher energy efficiency, and the stable Coulombic efficiency further verifies the outstanding cycling stability of this integrated configuration. Accordingly, appropriate PB loading effectively upgrades the overall output and energy conversion performance of neutral iron-sulfur flow batteries.

3.5.2. Energy Efficiency Variation of Neutral Iron-Sulfur Flow Batteries under Different Current Densities

The above comparative efficiency tests between PB-containing and blank full cells confirm that the improved energy efficiency mainly originates from suppressed voltage loss, with minor contributions from enhanced Coulombic efficiency. Energy efficiency serves as a comprehensive metric that integrates charge utilization, voltage hysteresis and overall energy conversion capability during practical operation. To further elucidate the cell performance responses under diverse operating conditions, energy efficiency was selected as the core evaluation index to analyze the influence of current density on the energy conversion behavior of neutral iron-sulfur flow batteries, with results illustrated in Figure 7.

As observed in the figure, under approximately 40 °C, the energy efficiency declines monotonically as the current density rises from 20 mA·cm⁻² to 60 mA·cm⁻². The corresponding energy efficiencies reach 89.23%, 86.58%, 85.21%, 80.67% and 75.96% at 20, 30, 40, 50 and 60 mA·cm⁻², respectively. This demonstrates that high energy conversion efficiency can be retained at low current densities, whereas aggravated internal polarization at elevated current rates deteriorates energy efficiency.

The decay trend varies across different current density intervals. A moderate reduction in energy efficiency is detected from 20 mA·cm⁻² to 40 mA·cm⁻², signifying well-matched charge-discharge processes and satisfactory energy utilization within this range. In contrast, a much steeper efficiency drop occurs above 50 mA·cm⁻², which arises from amplified activation polarization, ohmic polarization and concentration polarization at high rates.

These polarization effects widen the voltage gap between charging and discharging, resulting in substantial energy efficiency loss.

Such performance degradation at high current densities is fundamentally attributed to constrained reaction kinetics and mass transport. On one hand, elevated current densities demand accelerated electron transfer at electrode interfaces per unit time; mismatched interfacial reaction kinetics inevitably exacerbates electrode polarization. On the other hand, rapid consumption of redox species at electrode surfaces under high-rate operation causes insufficient replenishment via convection and diffusion in electrolyte, triggering severe concentration polarization and further enlarging voltage loss. For the system investigated herein, although optimized PB dosage improves the utilization of iron-based active materials, the aforementioned polarization losses cannot be fully eliminated under high current densities, leading to the gradual drop of energy efficiency with increasing current density.

In conclusion, neutral iron-sulfur flow batteries sustain favorable energy efficiency within the current density window of 20–40 $\text{mA} \cdot \text{cm}^{-2}$, while energy efficiency suffers dramatic deterioration above 50 $\text{mA} \cdot \text{cm}^{-2}$, revealing that high-rate operation imposes severe adverse impacts on the energy conversion performance of the system.

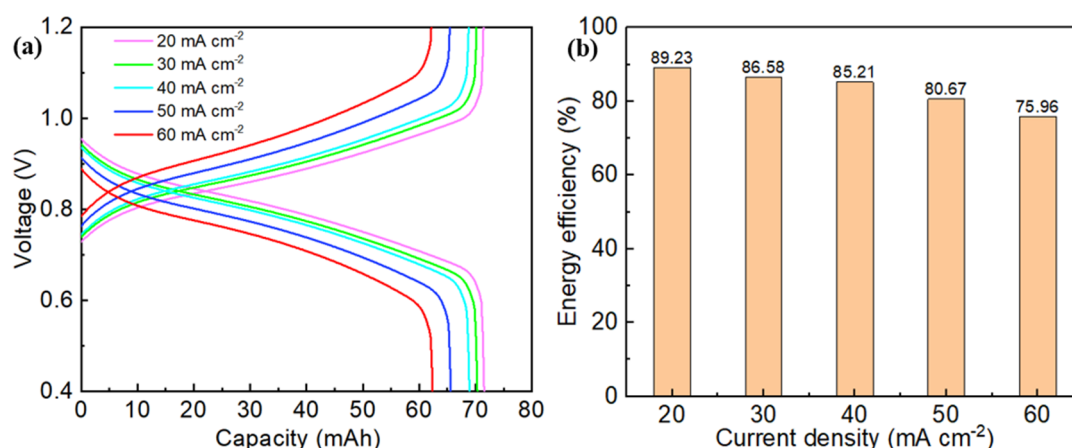


Figure 7. (a) Galvanostatic charge-discharge profiles and (b) energy efficiency plots of neutral iron-sulfur flow batteries under various current densities.

3.5.3. Energy Efficiency Variation of Neutral Iron-Sulfur Flow Batteries at Different Temperatures

To further explore the temperature dependence of energy conversion performance for neutral iron-sulfur flow batteries, energy efficiency measurements were carried out at 30, 40 and 50 $^{\circ}\text{C}$ under a current density of 30 $\text{mA} \cdot \text{cm}^{-2}$, and the corresponding results are displayed in Figure 8. The energy efficiency rises from 85.21% to 86.52% as the temperature increases from 30 $^{\circ}\text{C}$ to 40 $^{\circ}\text{C}$, whereas it decreases to 83.85% when the temperature is further elevated to 50 $^{\circ}\text{C}$. This reveals the dual effect of temperature on cell performance: moderate heating facilitates energy conversion efficiency, while excessively high temperature exerts negative impacts on the overall system performance.

Within the temperature range of 30–40 $^{\circ}\text{C}$, elevated temperature accelerates the diffusion of active species in electrolyte and the charge-transfer kinetics at electrode interfaces. Accordingly, the electron exchange between soluble $[\text{Fe}(\text{CN})_6]^{4-}/[\text{Fe}(\text{CN})_6]^{3-}$ mediators and solid PB is boosted, and the interfacial reaction kinetics of polysulfide species on the negative side are also optimized. Consequently, the cell achieves higher energy efficiency at 40 $^{\circ}\text{C}$, demonstrating that mild temperature rise mitigates reaction polarization and elevates the energy utilization efficiency of the battery system.

Nevertheless, energy efficiency drops distinctly at 50 $^{\circ}\text{C}$. Apart from universal drawbacks including polysulfide shuttling and cross-diffusion of active materials, this PB-containing system suffers two unique high-temperature side effects. Repeated potassium ion insertion/extraction under thermal stress induces lattice distortion and structural instability of solid PB, reducing the utilization of solid energy storage materials. Meanwhile, high temperature breaks Fe–CN coordination bonds and accelerates ferrocyanide hydrolysis. The hydrolysis reaction is particularly promoted at elevated temperatures, leading to irreversible loss of the redox mediator. Combined with unfavorable mass transfer behaviors, these two specific side reactions exacerbate voltage loss during cycling and decrease energy efficiency.

In summary, within the temperature range investigated in this work, the neutral iron-sulfur flow battery delivers the maximum energy efficiency at 40 $^{\circ}\text{C}$. This indicates that moderate temperature elevation can enhance the electrochemical reactivity and energy conversion performance of the system, whereas excessively high

temperatures are detrimental to stable cell operation. Accordingly, 40 °C is identified as the optimal operating temperature for the present system.

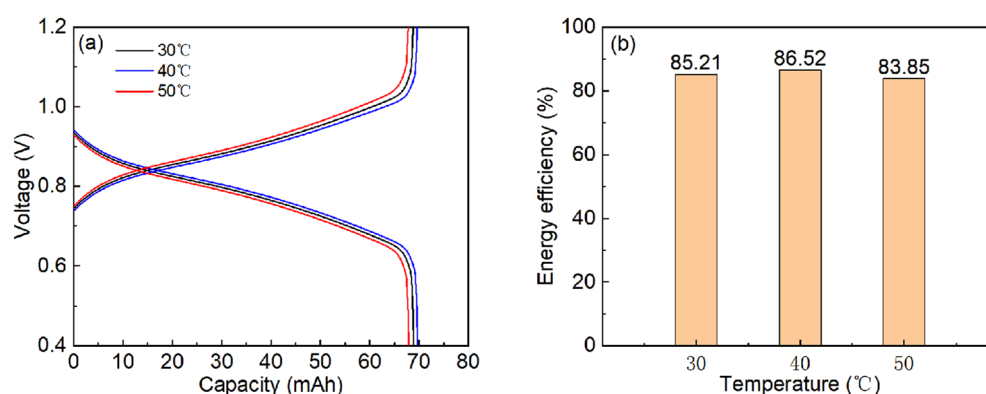


Figure 8. (a) Galvanostatic charge-discharge curves and (b) energy efficiency plots of neutral iron-sulfur flow batteries at different temperatures.

To contextualize the performance of the proposed neutral iron-sulfur flow battery, Table 2 compares its key electrochemical metrics with those of recently reported neutral or iron-based flow battery systems.

Table 2. Performance comparison of the proposed neutral iron-sulfur flow battery with recently reported neutral or iron-based flow battery systems.

Electrolyte/Strategy	CE (%)	EE (%)	Capacity/Energy Density	Reference
Near-neutral all-iron (Fe-racEDDHA/Fe(CN) ₆)	~100	~81.5	4.56 Ah L ⁻¹	Schröder et al. (2022) [36]
Neutral Zn-Fe (ZnBr ₂ /K ₃ Fe(CN) ₆)	~100	~60–70	3.2–26.9 Wh L ⁻¹	Yang et al. (2022)[37]
Neutral all-iron + FeOB (Fe ²⁺ -DTPA/Fe(CN) ₆)	~99	—	Increased by 1.37×	Li et al. (2024) [38]
Near-neutral Fe-Cr (Cr(EDTA)(2-MI)/Fe(CN) ₆)	97	76	—	Li et al. (2025) [39]
Neutral all-iron (Fe(EDTMP)/K ₄ Fe(CN) ₆)	~100	~79.2	31.67 Ah L ⁻¹	Xiao et al. (2026) [40]
Neutral Zn-Fe (D-Asp/ZnCl ₂ /Na ₄ Fe(CN) ₆)	99.81	70.64	20 mAh cm ⁻²	Wang et al. (2026) [41]
Neutral iron-sulfur + NiS _x -GF + PB redox-targeting	~98	86.5	69.66 mAh	This work

As shown in Table 2, early studies (2022) primarily focused on system feasibility verification, but were constrained by low solubility of active species and zinc dendrite formation, resulting in limited capacity and efficiency. From 2024 to 2025, strategies such as microbial incorporation and dual-ligand coordination were introduced to improve kinetics, albeit at the cost of increased system complexity. In 2026, the Fe(EDTMP) and D-Asp additive systems achieved notable progress in capacity and cycling performance, yet involved complex ligand synthesis or relatively high material costs. In contrast, the present work adopts a Prussian blue redox-targeting strategy and achieves an energy efficiency of 86.5% (obtained at 40 °C and 30 mA cm⁻²) and a discharge capacity of 69.66 mAh, while utilizing a low-cost, environmentally benign Fe/S/PB material system that combines competitive performance with practical application potential.

4. Conclusions

Addressing the critical limitation of insufficient positive-side capacity originating from the ferricyanide couple in neutral iron-sulfur flow batteries, this work carries out systematic research on positive capacity enhancement, followed by full-cell performance validation. The major conclusions are summarized as follows:

- (1) A redox-targeted symmetric flow battery system is constructed, where aqueous [Fe(CN)₆]⁴⁻/[Fe(CN)₆]³⁻ serves as the liquid redox mediator and PB acts as solid energy storage material. The activation behavior of PB and the effect of PB loading are comprehensively investigated. Within the tested ferricyanide concentration range of 0.1–0.3 M, the 0.3 M electrolyte enables remarkable capacity contribution from PB and is thereby selected as the representative working concentration for subsequent PB dosage optimization. Among the four tested PB molar ratios (10%, 20%, 30%, 40%), the 20% loading delivers balanced capacity increment and material utilization efficiency, which is determined as the optimal PB addition protocol under the experimental conditions of this work.

- (2) Full-cell measurements are performed by integrating PB with the optimized dosage into neutral iron-sulfur flow batteries. Experimental results demonstrate that with NiS_x-GF composite electrodes as the negative electrode, the full cell incorporated with optimal PB achieves a peak power density of 14.58 mW cm⁻² and a discharge capacity of 69.66 mAh, accompanied by substantially elevated energy efficiency. In addition, 40 °C is confirmed as the optimal operating temperature. These findings reveal that proper PB loading not only improves the deliverable capacity of the positive compartment but also alleviates overall cell polarization and promotes energy conversion efficiency.

In summary, this paper systematically explores performance improvement strategies for neutral iron-sulfur flow batteries from two perspectives: positive capacity reinforcement and full-cell validation. The results verify that PB with optimized loading can boost the capacity output of the iron-based positive electrode via redox-targeted reactions, and simultaneously upgrade the capacity, power density and energy efficiency of neutral iron-sulfur full cells. This work provides solid experimental support for further optimization of neutral iron-sulfur flow battery systems.

Although this paper has achieved certain progress in improving the performance of neutral iron-sulfur flow batteries, several issues remain to be further explored and investigated. Future researches should focus on the following aspects.

- (1) Further refine the key influencing factors of the capacity enhancement process via Prussian blue redox-targeted reactions. This study has screened the optimal ferricyanide working concentration and PB dosage under the given experimental conditions, while their applicable scopes are still limited by the test parameters. Subsequent research can conduct in-depth and detailed investigations on multiple factors, including ferricyanide concentration, PB particle size, dispersion/fixation state of particles in the electrolyte reservoir, solid-liquid contact mode, and reservoir structure. Such efforts are expected to further enhance the utilization efficiency of PB and accurately define the effective capacity enhancement range of the redox-targeted strategy.
- (2) Further explore the long-term cycling stability and performance decay mechanism of full batteries. This paper mainly focuses on full-cell performance validation and performance analysis under typical operating conditions. However, in-depth investigations are still required on key issues during long-term cycling, such as membrane fouling, cross-diffusion of active species, polysulfide shuttling, structural stability evolution of PB, and electrode surface deactivation. Combined with material characterization and failure analysis before and after cycling, future work can effectively reveal the intrinsic sources of performance decay, providing theoretical and experimental support for the stability improvement of neutral iron-sulfur flow battery systems.
- (3) The incorporation of solid Prussian Blue particles creates multiple engineering challenges for large-scale practical deployment, and targeted research on process optimization should be conducted in future studies. For one thing, solid PB tends to aggregate after long-term standing in electrolyte, which significantly reduces solid-liquid contact efficiency and impairs the capacity enhancement effect of redox targeting. For another, the degraded fluidity of solid-liquid mixed electrolyte slurry increases energy consumption for circulating pumping. Further optimization research focusing on the above issues can be implemented to advance the practical large-scale energy storage application of this redox-targeted capacity-boosting strategy.

Author Contributions

J.Z. and Y.Q.: Conceptualization; Writing—original draft; X.H.: Investigation; Z.L., H.S. and P.L.: Supervision; Q.M.: Funding acquisition; Supervision; Q.X.: Conceptualization; Funding acquisition; Writing—review & editing; Supervision. All authors have read and agreed to the published version of the manuscript.

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Institutional Review Board Statement

Not applicable.

Data Availability Statement

The raw data that support the findings of this study are available from the corresponding author upon reasonable request.

Conflicts of Interest

The authors declare no conflict of interest.

Use of AI and AI-Assisted Technologies

No AI tools were utilized for this paper.

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