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A mini review on key properties and requirements of vanadium electrolytes

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Abstract. This article examines the key factors involved in the production of electrolytes for vanadium batteries. It delves into various important parameters, including the optimal concentration of vanadium in the electrolyte, the composition of the electrolyte itself, and the necessity of maintaining appropriate levels of ionic strength, viscosity, temperature, and electrical conductivity in the solution. Moreover, the article highlights the significance of preventing electrode wear and sediment formation, offering insights into different reagents used and their specific mechanisms of action. By providing such valuable information, this article emphasizes the utmost importance of manufacturing high-quality vanadium batteries and their essential electrolytes, especially considering the rapid advancements in this field. This comprehensive study sheds light on the crucial parameters that must be carefully considered during the production of electrolytes for vanadium batteries. It stresses the criticality of maintaining optimal concentrations, proper composition, and suitable levels of ionic strength, viscosity, temperature, and electrical conductivity. It is also underscoring the need to prevent electrode wear and sediment formation. Generally speaking, this article serves as an invaluable resource in the production of top-notch vanadium batteries and their indispensable electrolytes in light of the rapid progress in this domain.

Keywords: *vanadium batteries, electrolyte concentration, electrolyte viscosity, redox processes, electrodes.*

1. Introduction

Vanadium batteries have gained significant attention in recent years as a promising energy storage solution for renewable energy sources [1-3]. The ability to store and release energy efficiently is crucial for the integration of intermittent renewable energy sources into the electricity grid. Vanadium batteries, particularly the vanadium redox flow batteries (VRFBs), have emerged as a viable option due to their scalable design, long cycle life, and high energy efficiency. Central to the functioning of these batteries is the electrolyte composition, which plays a crucial role in determining their performance and overall efficiency [4].

Research efforts have been focused on improving the composition of electrolytes to enhance the storage capacity, charging efficiency, and stability of vanadium batteries. Electrolytes are critical components as they enable the flow of ions between the positive and negative electrodes during the charge and discharge processes. The choice of electrolyte composition affects the battery's performance, energy density, and overall efficiency. Thus, understanding and improving the electrolyte composition is of paramount importance for the development of more efficient vanadium batteries.

Several studies have investigated the effects of different electrolytes on the performance of vanadium batteries. For instance, Puleston et al. [5] conducted a comprehensive review of the electrolyte compositions used in VRFBs and highlighted the significance of optimizing the concentration and composition of vanadium species. They emphasized the need for developing high-performance electrolytes with improved stability and higher energy density. Despite the growing interest in vanadium batteries and their electrolytes, there are still gaps in our knowledge. One area that requires further research is the development of low-cost and sustainable electrolyte solutions for large-scale applications. Traditional VRFBs utilize expensive and environmentally hazardous vanadium salts as electrolytes. However, recent studies have explored alternative electrolytes, such as organic compounds and non-vanadium redox couples. These alternative electrolytes offer the possibility of reducing costs, enhancing sustainability, and expanding the range of applications for vanadium batteries [6].

The schematic structure of vanadium batteries (Figure 1), specifically vanadium redox flow batteries (VRFBs), consists of several key components that enable their operation. These components include the electrolyte tanks, the stack of electrochemical cells, the flow loop, and the power management system. Understanding the principle of operation of VRFBs is crucial for comprehending their potential in energy storage applications.

At the heart of VRFBs are two separate electrolyte tanks, typically referred to as the positive electrolyte tank and the negative electrolyte tank. These tanks contain vanadium-based electrolyte solutions with different redox states. The positive electrolyte tank houses the positive electrolyte solu-

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tion with vanadium ions in the +5 oxidation state (V5+), while the negative electrolyte tank contains the negative electrolyte solution with vanadium ions in the (+2) oxidation state (V2+). The distinction in redox states of the electrolytes is one of the distinguishing features of VRFBs [1,7].

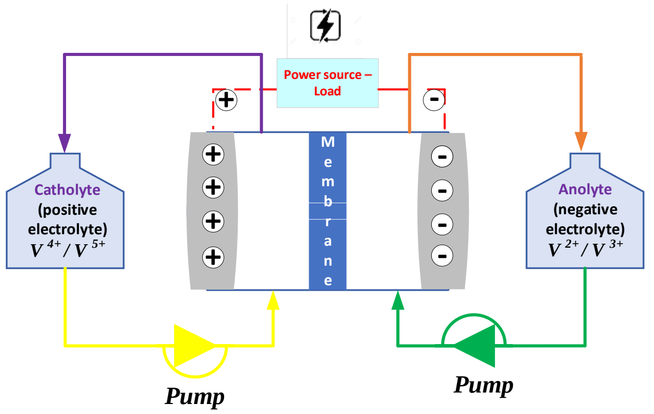

Figure 1. Schematic representation of vanadium batteries - redrawn from [1]

The electrochemical cells of a VRFB stack are the main units responsible for converting chemical energy to electrical energy and vice versa. Each cell consists of a positive and a negative electrode, separated by a membrane. The electrodes are typically made of carbon-based materials that provide a large surface area for the exchange of ions. The membrane, often made of an ion-conducting polymer, allows the selective passage of vanadium ions while preventing the crossover of other species [8].

During the charge process, the electrolytes flow through the stack of electrochemical cells in a continuous manner. The negatively charged vanadium ions (V^{2+}) from the negative electrolyte solution are oxidized at the positive electrode, releasing electrons and transforming into positively charged vanadium ions (V^{3+}). These electrons flow through the external circuit, providing electrical energy. Meanwhile, the positively charged vanadium ions (V^{5+}) in the positive electrolyte solution are reduced at the negative electrode, accepting the electrons and transforming into negatively charged vanadium ions (V^{4+}). Conversely, during the discharge process, the flow of electrolytes through the electrochemical cells is reversed. The negatively charged vanadium ions (V^{4+}) from the positive electrolyte solution are oxidized at the negative electrode, releasing electrons and transforming into positively charged vanadium ions (V^{5+}). These electrons again flow through the external circuit to provide electrical energy. Meanwhile, the positively charged vanadium ions (V^{3+}) in the negative electrolyte solution are reduced at the positive electrode, accepting the electrons and transforming into negatively charged vanadium ions (V^{2+}). The overall reaction can be represented as follow:

$$VO^{2+} + V^{3+} + H_2O \rightarrow VO_2^{+} + V^{2+} + 2H^{+} \quad (1)$$

As the electrolytes flow through the cell stack, the vanadium ions undergo continuous oxidation and reduction reactions, enabling the storage and release of electrical energy. The flow loop, consisting of pumps and pipes, facilitates the circulation of the electrolytes between the electrolyte tanks and the stack of electrochemical cells. This flow-based design allows for flexible scalability and long cycle life [9].

The power management system of VRFBs controls the charging and discharging processes, ensuring efficient operation and preventing overcharging or overdischarging. It monitors and adjusts the flow rates of the electrolytes based on energy demand and manages the potential imbalances between the positive and negative electrolyte solutions.

2. Materials and methods

2.1. The essential requirements for the concentration of vanadium electrolyte in the usage of vanadium batteries

Figure 2 illustrates the increasing research efforts aimed at addressing the challenges associated with the concentration of vanadium electrolyte in vanadium batteries. These batteries are gaining popularity in various industries due to their high energy density and long service life. However, the strict requirements for the concentration of vanadium electrolyte pose a significant obstacle to their widespread use. Improving the conditions and properties of the electrolyte in vanadium batteries is crucial for further enhancing their existing advantages. Therefore, research work in this area is on the rise, with the aim of overcoming the problems and limitations associated with vanadium batteries and their electrolytes.

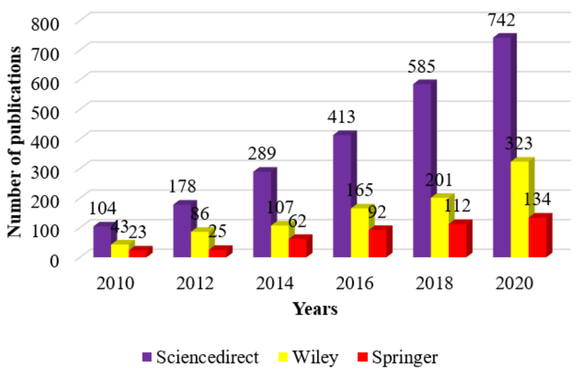

Figure 2. Scientific publications published from 2010 until mid-2020 on vanadium batteries - redrawn from [10]

Extensive research has established that the optimal concentration of vanadium in the electrolyte falls within the range of 1.5 to 2.0 m (molarity) [11,12]. Within this range, vanadium batteries exhibit the most favorable combination of energy density, specific power, and service life.

However, issues may arise if the concentration of vanadium electrolyte exceeds the recommended range. Primarily, a higher concentration elevates the viscosity of the electrolyte, impeding the movement of vanadium ions within the battery. This results in increased internal resistance and reduced overall efficiency. Moreover, utilizing a highly viscous solution necessitates additional operational requirements, consequently raising the costs associated with vanadium battery applications. Another drawback of excessive vanadium concentration in the solution is the formation of solid sludge or sediment-like precipitates within the battery. The formation of such a precipitate creates blockages in the flow channels within the battery, hindering the movement of vanadium ions. As a consequence, the battery's efficiency may decrease, and the functioning of the operational systems can be compromised [13].

Optimal performance of the battery necessitates maintaining the concentration of vanadium electrolyte within the recommended range. Insufficient concentration results in diminished energy density and restricts the battery's energy storage capacity. Additionally, a low vanadium concentration can result in decreased voltage, rendering the battery unsuitable for high-power applications. Insufficient electrolyte concentration also elevates the self-discharge rate of the battery, reducing its ability to retain stored energy over an extended period.

To emphasize the significance of maintaining the optimal concentration of vanadium electrolyte, let us consider an example. Suppose the concentration of vanadium in the battery electrolyte exceeds the recommended range. As a result of increased viscosity, the internal resistance of the battery rises, causing substantial energy losses during charge and discharge cycles. This decrease in overall efficiency leads to reduced operating time and diminished energy production. Furthermore, the elevated concentration can result in the formation of a precipitate within the battery, obstructing the movement of vanadium ions and clogging the flow channels. These negative effects significantly curtail the energy capacity of the battery, consequently shortening its lifespan [14].

Conversely, if the concentration of vanadium electrolyte falls below the specified effective range, the energy storage capacity of the battery will be diminished. Deviations in the vanadium electrolyte concentration from the recommended range can give rise to various issues, including increased electrolyte viscosity, sediment formation at the bottom of the battery, and decreased specific energy and power. Therefore, meticulous attention must be given to maintaining the appropriate concentration of vanadium electrolyte to ensure the efficiency and optimal functioning of batteries utilizing vanadium electrolytes.

2.2. The fundamental operational principle of vanadium batteries

Vanadium batteries consist of two cells: one with a low oxidation state electrolyte containing vanadium ions (V^{2+} and V^{3+}), and the other with a high oxidation state electrolyte containing vanadium ions (V^{4+} and V^{5+}). Maintaining the correct balance of these oxidation states is crucial for the long-term lifespan and overall efficiency of the battery.

The electrical energy in vanadium batteries is stored as chemical potential energy through reversible redox reactions at the positive and negative electrodes. During charging, electrons from an external power source convert V^{2+} ions in the negative electrolyte to V^{3+} ions, and V^{5+} ions in the positive electrolyte to V^{4+} ions. During discharge, these redox reactions are reversed, converting V^{3+} ions back to V^{2+} ions in the negative electrolyte and V^{4+} ions back to V^{5+} ions in the positive electrolyte. The ability of vanadium batteries to store and deliver electricity depends on redox reactions involving vanadium ions of different oxidation states. Therefore, maintaining the balance of these reactions is vital for the overall performance and cost-effectiveness of these batteries. Imbalances in the oxidation state balance can lead to capacity loss, reduced efficiency, and battery failure [15].

One of the key factors affecting the balance of oxidation states is the transfer of vanadium ions between the positive and negative electrolytes. This transfer occurs through an ion-selective membrane during a transition period. However, the use of low-quality diaphragm materials can disrupt the oxidation state balance and decrease performance. This chaotic migration of vanadium ions between electrolytes is termed cross-movement. To address this issue, research is focused on using highly selective ion-selective membranes, such as Naphion, that prevent the chaotic mixing of vanadium ions by selectively transporting protons.

Another research [16] focus is optimizing the composition and concentration of electrolyte solutions to maintain the balance of vanadium ion oxidation states. Various additives can be incorporated to improve the selectivity of ion-selective membranes and reduce the negative mutual effects of vanadium ions. The design and configuration of vanadium batteries also play a role in oxidation state balance. Factors like electrode area, geometry, and electrolyte flow rate influence redox reactions and vanadium ion migration. Thus, correctly shaping the electrodes and controlling electrolyte flow aid in maintaining the proper distribution of vanadium ions in the solution. Other factors such as temperature, pH, and electrolyte composition can also impact redox reactions and vanadium ion migration. Therefore, managing and optimizing these operational parameters are crucial for achieving optimal battery performance.

Furthermore, the use of advanced materials and technologies can enhance battery efficiency and performance. Researchers are exploring the utilization of various electrode materials, such as carbon-based materials and metal oxides, to improve redox reactions and increase energy density. Additionally, the development of new ion-selective membranes with enhanced selectivity and stability can further enhance electrolyte balance and overall battery performance.

3. Results and discussion

3.1. Effect of ionic strength

The ionic strength is a critical factor in the utilization of vanadium redox batteries, directly impacting their efficiency and performance. It plays a vital role in determining the overall effectiveness of these batteries.

Firstly, the ionic strength significantly influences the conductivity of the electrolyte solution. Higher ionic strength results in increased conductivity, enhancing the overall efficiency of the battery. This improved conductivity facilitates faster transport of ions between the electrodes, reducing internal resistance and voltage losses. Additionally, higher ionic strength enhances the solubility of vanadium ions, which is crucial for maintaining the power and energy density of the battery. Insufficient solubility can lead to the formation of deposits on the electrodes, compromising the performance and overall integrity of the battery. Furthermore, the electrochemical reactions occurring at the electrodes are also impacted by the ionic strength. The speed of redox reactions involving vanadium ions is directly related to the ionic strength of the electrolyte. High ionic strength promotes faster charging rates for the battery. However, it is important to note that excessively high ionic strength can have detrimental effects on battery performance [17].

Elevated ionic strength can increase viscosity and reduce ion movement (diffusivity), ultimately slowing down the kinetics of redox reactions. Therefore, ongoing research is focused on finding the optimal balance of ionic strength that maximizes battery performance without compromising its overall efficiency.

3.2 Effect of conductivity of the electrolyte

Ionic conductivity refers to the capacity of an electrolyte to facilitate the flow of electric current via ion movement. In the case of vanadium batteries, the electrolyte solution comprises vanadium ions in different oxidation states (V^{2+}/V^{3+} and V^{4+}/V^{5+}). To ensure optimal conductivity, a strong acid, typically sulfuric acid, is commonly employed. As the movement of vanadium ions within the electrolyte greatly impacts the power output, efficiency, and overall performance of the battery, it is a crucial factor in the functionality of vanadium batteries [7].

The pace at which vanadium ions move through the electrolyte solution hinges on the ionic conductivity of the electrolyte. Higher conductivity translates into swifter ion mobility, resulting in amplified power generation and efficiency for vanadium batteries. Numerous elements influence conductivity, including the concentration of vanadium ions, temperature, electrolyte viscosity, and the presence of impurities in the electrolyte solution [1,18].

3.3 Effect of vanadium concentration

Raising the concentration of vanadium ions enhances ionic conductivity by providing additional ions for the conduction of electric current. However, exceedingly high concentrations can result in heightened viscosity, which inhibits the movement of ions and leads to a decline in ionic conductivity. The figure 3 clearly demonstrates a strong correlation between the concentration of vanadium in solution and its electrical conductivity.

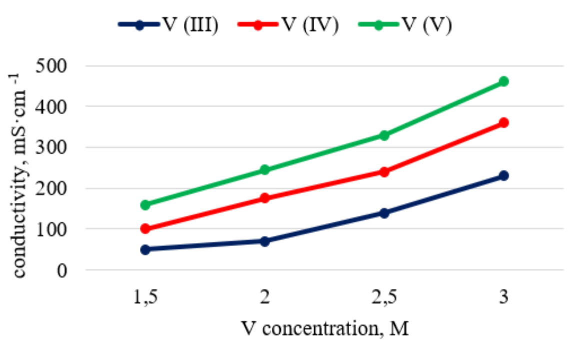

Figure 3. Conductivity due to different oxidation states of vanadium at 20 ºC [19]

This relationship is supported by scientific evidence and can be explained by the oxidation states of vanadium. When comparing different degrees of oxidation of vanadium, it becomes evident that a higher degree of oxidation is associated with a higher electrical conductivity. This can be attributed to the alteration in electronic structure and charge distribution within the vanadium ions as they undergo oxidation. As the oxidation state of vanadium increases, so does the number of available valence electrons, enabling a higher mobility of charge carriers within the electrolyte. A concrete example of this phenomenon is observed when comparing vanadium electrolytes with different concentrations of vanadium ions. A higher concentration of vanadium ions results in a larger number of vanadium atoms available for oxidation and consequent increase in electrical conductivity. This can be visually represented in the figure, where higher concentrations are clearly associated with higher electrical conductivity values.

3.4 Effect of temperature

Temperature is a crucial factor in influencing electrolyte conductivity. Elevated temperatures generally enhance ion mobility, thereby increasing ionic conductivity. This occurs because higher temperatures provide the ions with greater kinetic energy to overcome electrostatic forces, allowing them to move more freely. However, excessive temperatures can lead to thermal degradation of the electrolyte and a subsequent decrease in battery performance (Table 1).

Table 1. The effect of electrolyte temperature on voltage efficiency and electrical conductivity [20]

№	Temperature, ºC	Voltage efficiency, (%)	Conductivity, $mS\cdot cm^{-1}$
1	15	86,5	430
2	25	87,6	480
3	35	88,4	525
4	45	89,2	560
5	55	90,5	620

The rise in temperature induces an increase in kinetic energy, causing the movement of charged particles within the electrolyte to accelerate. This accelerated motion leads to more collisions between the charge carriers, resulting in enhanced ion mobility and increased electrical conductivity. The relationship between temperature and electrical conductivity can be mathematically described by the Arrhenius equation. According to this equation, the electrical conductivity (σ) of the electrolyte increases exponentially with increasing temperature (T), following the equation:

$$\sigma = \sigma_0 \cdot \exp(Ea/RT) \quad (2)$$

Where σ_0 - represents the pre-exponential factor, Ea - the activation energy barrier, R - is the ideal gas constant, T - represents the absolute temperature.

From this equation, it is evident that an increase in temperature leads to a higher value for σ. The improved electrical conductivity at higher temperatures has several benefits for vanadium batteries. Firstly, it results in reduced internal resistance, allowing for more efficient charge transfer between the vanadium ions during the redox reactions. This increased efficiency translates into higher energy conversion and storage capabilities. Additionally, the enhanced electrical conductivity promotes better electrochemical kinetics and higher power output, making vanadium batteries more suitable for demanding applications that require rapid energy transfer.

However, it is important to note that excessively high temperatures can have detrimental effects on the overall performance and lifespan of vanadium batteries. Extreme temperatures can cause accelerated degradation of the electrolyte and electrode materials, leading to reduced capacity and decreased cycle life.

3.5 Effect of viscosity

Viscosity is a significant factor influencing the ionic conductivity of electrolytes. It refers to the internal resistance of a liquid. An increase in the viscosity of the electrolyte results in greater resistance for ions to move through the solution, leading to a decrease in ionic conductivity. Thus, maintaining an optimal range of viscosity is crucial to ensure adequate ion transport and efficient operation of vanadium batteries [20,21].

The presence of impurities in the electrolyte can also impact ionic conductivity. However, certain additives can increase ionic conductivity by enhancing the solubility and mobility of vanadium ions. These additives may include organic compounds, surfactants, or salts that can modify the properties of the electrolyte and enhance its ionic conductivity. In recent years, researchers have been exploring various methods and materials to enhance the ionic conductivity of electrolytes in vanadium batteries. Through their work, new electrolyte compositions have been developed that exhibit low viscosity and high ionic conductivity, without compromising the essential properties required for battery operation. For instance, researchers have investigated the addition of phosphoric acid and acetic acid to electrolytes, which has been found to increase their ionic conductivity and improve overall battery performance [22]. In addition to acid concentration, the vanadium content in the solution can also influence the viscosity of the electrolyte. For instance, as mentioned in article [23], the authors assert that an increase in vanadium concentration leads to a corresponding rise in the electrolyte's viscosity (Figure 4).

Overall, research efforts are focused on optimizing the composition and properties of electrolytes to enhance the ionic conductivity and efficiency of vanadium batteries.

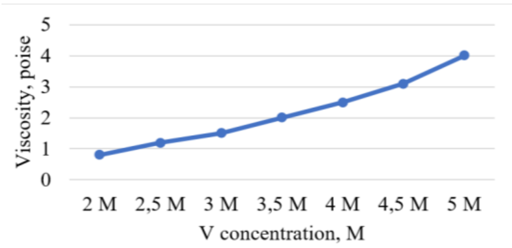

Figure 4. The effect of vanadium concentration on the electrolyte viscosity [23]

One area of research with promising results focuses on the development of ion-permeable membranes for use in vanadium batteries. These membranes are designed to separate the positive and negative electrolyte solutions, enabling the movement of vanadium ions. Researchers [24] are working to optimize the properties of these membranes, such as porosity, thickness, and chemical composition, in order to increase the ionic conductivity and improve the overall efficiency of vanadium batteries.

Another study has found that the incorporation of nanomaterials in vanadium batteries can enhance the ionic conductivity of the electrolytes. Nanomaterials such as carbon nanotubes, graphene, and metal oxides can be added to the electrolyte solution or used as additives to improve the transport properties of ions. These nanomaterials provide a large surface area for ion adsorption, leading to an increased rate of ion diffusion. This, in turn, improves the ion conductivity capacity and overall performance of vanadium batteries.

4. Conclusions

The review article presents a meticulous analysis of the requirements for vanadium electrolytes in batteries, consolidating data from various published works. The article extensively covers significant aspects related to the operation of vanadium batteries, including the impact of ionic force, vanadium concentration, temperature, and viscosity on battery performance. The findings highlight the crucial role played by these parameters in determining the efficiency and functionality of vanadium batteries. The article provides essential data on the optimal values for each of these variables, enabling researchers and industry professionals to enhance the overall performance of vanadium batteries. By synthesizing a multitude of research papers, this review article assumes a unique position as a valuable source of information. It serves as a comprehensive reference for individuals seeking to expand their knowledge and understanding of vanadium batteries.

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Ванадий электролиттерінің негізгі қасиеттері және оларға қойылатын талаптар бойынша шағын шолу

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Аңдатпа. Мақалада ванадий батареяларында қолданылатын электролиттерді өндіруге қадағаланатын негізгі факторлар талданған. Электролиттер өндірісі барысында сақаталатын параметрлерді, соның ішінде электролиттегі ванадийдің тиімді концентрациясын, электролиттің құрамын және ерітіндідегі иондық беріктіктің, тұтқырлықтың, температураның және электр өткізгіштіктің тиісті деңгейлерін сақтау қажеттілігін айқындайды. Сонымен қатар, мақалада электродтардың тозуын және шөгінділердің пайда болуын болдырмаудың маңыздылығы көрсетіліп, осы мақсатта қолданылатын әртүрлі реагенттер және олардың әсер ету механизмдері туралы түсінік берілген. Осындай құнды ақпаратты қамтыған мақалада жоғары сапалы ванадий батареялары мен олардың маңызды электролиттерін өндірудің, әсіресе осы саладағы қарқынды жетістіктерді ескере отырып, осы өндірістің маңыздылығына баса назар аударылады. Бұл жан-жақты әдеби зерттеу жұмысы ванадий батареяларына арналған электролиттерді өндіру кезінде мұқият қарастырылуы керек маңызды параметрлерді анықтайды.Сонымен қатар мақалада электродтардың тозуын және шөгінділердің пайда болуын болдырмау жолында жасалатын іс-шаралар туралы негізгі процестер сипатталған. Жалпы айтқанда, бұл мақала осы саладағы қарқынды прогрестерді ескере отырып, жоғары сапалы ванадий батареяларын және олардың электролиттерін өндіруде құнды ресурс ретінде қызмет етеді.

Негізгі сөздер: *ванадий батареялары, электролит концентрациясы, электролит тұтқырлығы, тотығу-тотықсыздану процестері, электродтар.*

Мини-обзор основных свойств и требований к ванадиевым электролитам

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Аннотация. В статье рассматриваются ключевые факторы, связанные с производством электролитов для ванадиевых аккумуляторов. В нем рассматриваются различные важные параметры, включая оптимальную концентрацию ванадия в электролите, состав самого электролита и необходимость поддержания соответствующих уровней ионной силы, вязкости, температуры и электропроводности в растворе. Кроме того, в статье даны методы предотвращения износа электродов и образования осадка, дается представление о различных используемых реагентах и их специфических механизмах действия. Предоставляя вышеописанную информацию, эта статья подчеркивает исключительную важность производства высококачественных ванадиевых аккумуляторов и их основных электролитов, особенно учитывая стремительный прогресс в этой области. Это исследование охватывает важнейшие параметры, которые необходимо тщательно учитывать при производстве электролитов для ванадиевых аккумуляторов. В нем проиллюстрировано важность поддержания оптимальных концентраций, надлежащего состава и подходящих уровней ионной силы, вязкости, температуры и электропроводности. Также подчеркивается необходимость предотвращения износа электродов и образования осадка. В общем, статья является значимым ресурсом информации для производства ванадиевых аккумуляторов и их неотъемлемых электролитов, особенно учитывая быстрый прогресс в данной сфере.

Ключевые слова: *ванадиевые батареи, концентрация электролита, вязкость электролита, окислительно-восстановительные процессы, электроды.*

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