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ENERGY STORAGE WILEY

Are biologically synthesized electrolytes the future in green energy storage?

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Abstract

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The production of renewable energy from solar panels and windmills is rapidly increasing these years. However, one of the biggest hurdles that need to be overcome in the green transition is cost-efficient energy storage to reach the full exploitation of their potential. A promising energy storage technology is redox flow batteries (RFBs), particularly using quinones as electron carriers. The prototypes of quinone batteries have been derived from crude oil, which unfortunately falls short of the ambition of sustainable energy production purely from renewable sources. It is well-known that filamentous fungi have a great capacity for quinone production. and the first RFB with a fungalproduced quinone was recently generated as a proof-of-concept. Here, we give our opinion and perspectives on which challenges need to be solved before an RFB with fungal-produced quinones can be applied in the green transition.

KEYWORDS

energy storage, fungi, polyketides, quinone, redox flow battery

1 | INTRODUCTION

The intermittency of renewable solar energy and wind power leads to periods with both high and low energy production during the day, which can vary up to 80% within time scales of minutes to hours.¹ This supports the need for storage technologies even more since the energy demand and consumption do not match this variability of available energy from renewables. The availability of surplus energy at the peak production does not always match the demand, resulting in surplus energy in the peak production periods as well as a lack of energy when the energy demand exceeds the energy production.^{2,3}

The traditional lithium-ion batteries are commonly used in, for example, electronics and transportation industries due to their high energy density, lightweight, low self-discharge, and high efficiencies. However, they are not suitable for long-duration grid-scale energy storage technology as they do not feature the possibility to cost-effectively store enough renewable energy required to facilitate the long discharge durations needed for regulating the intermittency of renewable energy sources.^{4,5}

The redox flow battery (RFB; Figure 1) is another viable, cost-effective, and safe technology that has been recognized for large-scale stationary energy storage, and offers energy and power to be scaled independently, as well as short response times (depending on the electrolyte kinetics).⁵⁻⁹ The RFB technology could be used to match the energy supply from renewables to the demand since the technology, in contrast to lithium-ion batteries, offers the ability of long-duration discharge for example, due to the ability to decouple energy and power as well as lower self-discharge ratios, which is a result of having the electrolytes stored in separate reservoirs.^{5,10-13}

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FIGURE 1 Schematic of a redox flow battery cell, containing an electrochemical cell and two separate reservoirs for the electrolytes. The two half-cells are separated by a membrane and the current collectors are connected to the power source/load. Solid arrows indicate the charging process, while dashed arrows indicate the discharging process. The small arrows indicate the flow direction.

Flexibility in the energy and power ratio is an advantage as the energy density refers to the energy possible to store per unit volume or mass whereas the power density refers to the energy transfer rate possible per unit volume or mass, thereby making it possible to design the RFB devise for the specific needs.¹⁴

Many chemistries have already been investigated and studied as the active material for RFB electrolytes. To date, the vanadium flow battery (VFB) is the most researched and commercialized RFB system¹¹ since it can be operated at high concentrations, offers long cycle life, high-power density, fast charge and discharge, quick response times^{15,16} as well as limited electrolyte crossover due to the use of closely related vanadium complexes as electrolytes. These consist of the VO_2^+/VO^{2+} couple $(V^{5+}/V^{4+}$ oxidation state) as the posolyte and the V^{3+}/V^{2+} couple $(V^{3+}/V^{2+}$ oxidation state) as the negolyte both dissolved in a sulfuric acid electrolyte.¹⁷⁻²⁰ VFBs allow a lifespan of 15 000 to 20 000 charge/discharge cycles²¹ and the literature even suggests that 270 000 charge–discharge cycles have been demonstrated.^{22,23}

2 | QUINONE RFBs

The use of water-soluble organic redox couples to constitute the positive and negative electrolyte solutions in

RFBs has been investigated since the 2010s.²⁴⁻²⁶ Among some of the organic compounds researched are quinones, viologen, TEMPO (2,2,6,6-tetramethylpiperidine-1-oxyl), and ferrocenes.²⁴ The organic compounds show comparable volumetric capacities with that of the vanadium species and are even higher in some cases. The volumetric capacity possible to obtain is directly linked to the solubility of the compound in the used solvent.¹³ Quinones consist of aromatic ring structures (benzene rings) linked with carbonyl groups. They are divided according to the number of benzene rings in the backbone structure.^{27,28} Having the simplest structure, benzoquinones consist of one aromatic ring with two carbonyl groups placed in either ortho- or para-position, while naphthoquinones are bis-cyclic aromatic and anthraquinones are multicyclic aromatic hydrocarbons.^{29,30}

In an aqueous solution, quinones undergo reversible two-electron reactions due to them having two carbonyl groups present in their structures.^{30,31} The redox properties of the quinones largely depend on the operating conditions such as the supportive electrolyte and pH. At low pH, the reduction reaction proceeds as a single-step twoelectron two-proton process. At high pH, the reduction reaction is a two-electron process and does not involve protons. In a neutral pH solution, the reduction reaction is somewhere in between and proceeds as a one-proton two-electron reaction or proceeds as seen in a high alkaline solution.^{6,32} Furthermore, the reduction potential varies with pH and becomes more negative with increasing pH. Since a low reduction potential of the negolyte is preferred to achieve high cell voltage of the RFB, alkaline supportive electrolytes are often used.³³

Quinones are well-researched in this field, and some performance characteristics seen from RFB full-cell cycling experiments in the literature with different quinones as electrolyte material can be seen in Figure 2. As seen in the figure, especially anthraquinone derivatives have been investigated over the years, and they show good solubility in both acidic solutions through sulfonation and in alkaline solutions through hydroxylation. Because of the low redox potentials observed for many of them, they are good candidates for negolyte materials.^{37,49} The anthraquinones also show better cycling stability compared to benzoquinones, which can be reasoned by greater chemical stability due to the additional two rings.

By using different synthetic approaches, the stability of anthraquinones has been improved remarkably and research in the anthraquinones such as 2,6-DBEAQ, 2,6-DPPEAQ, and DBAQ have shown good long-term stability with a capacity fade rate in the range of 8×10^{-3} to 4×10^{-2} %/day (Figure 2), and the fade rate has even been reported as low as 1.8×10^{-3} %/day when DPivO-HAQ was cycled against ferrocyanide.⁴⁴ Despite the good



FIGURE 2 Summary of quinone stability vs theoretical capacity obtained from the RFB cycling. ¹DMBQ³⁴; ²DMOBQ³⁴; ³DHDMBS³⁵; ⁴DHBQ³⁶; ⁶Bislawsone³⁷; ⁷2,3-HCNQ³⁸; ⁸NQSO³³; ⁹2,6-DBEAQ⁵; ¹⁰2,6-DPPEAQ³⁹; ¹¹2,6-N-TSAQ¹⁶; ¹²PEGAQ⁴⁰; ¹³2,6-DHAQ⁴¹; ¹⁴2,3-DHAQ⁴²; ¹⁵1,8-BDPAQCL₂⁴³; ¹⁶DPivOHAQ⁴⁴; ¹⁷DBAQ⁴⁴; ¹⁸DCDHAQ⁴⁵; ¹⁹AQDP⁴⁶; ²¹DAEAQ⁴⁷; ²²1,3,5,7-THAQ⁴⁸

long-term stability reported for these anthraquinones, the high synthetic cost still hinders their use in large-scale applications.¹⁶

Lowering the cost of the electrolyte material is of huge importance for the scalability of quinone RFBs. Most of the quinones tested as electrolytes in RFBs are synthetically produced from nonrenewable feedstocks and the cost of these organic compounds mainly relates to the complexity and scale of the synthesis.^{24,50} Crude oil is rich in anthracene, which can be oxidized on large scale to anthraquinone. Raw components from coal tar are also used on large scale to synthesize naphtho- and anthraquinones. But relying on the production of electrolytes from fossil-derived hydrocarbon sources does not suit as a sustainable solution due to the large volume of electrolytes needed for energy storage.⁵¹ If RFBs with quinone electrolytes should be considered as the future storage technology for renewable energy sources, the quinones should be highly available and of low cost. Therefore, sustainable and cheap production with the low environmental impact of the quinones is highly preferred.^{3,52}

3 | BIOSYNTHESIZED QUINONES—CHALLENGES AND PROSPECTS

A promising source of suitable cheaply produced quinones is the fungal kingdom, where more than ENERGY STORAGE _WILEY 3 of 7



FIGURE 3 A, Overview of the phoenicin gene cluster and biosynthetic pathway. B, Scematic representation of the redox reaction mechanism of phoenicin under alkaline conditions.⁵⁷

300 quinones of fungal origin have been described.^{29,53-55} The biosynthesis of fungal quinones is initiated by polyketide synthases (PKSs), which are multidomain enzymes that perform iterative condensation reactions of activated acetate and malonate units.⁵⁶ The resulting products can then be further modified by oxygenation, acylation, and cyclization by enzymes which are encoded by genes located adjacent to the core PKS gene in tightly packed gene clusters (Figure 3A).⁵⁸

Recently, the fungal biosynthesized bibenzoquinone phoenicin (Figure 3B) was used as a negolyte in combination with ferrocyanide to demonstrate proof-of-concept in an RFB.⁵⁷ The resulting battery had a capacity of 11.75 Ah/L, which is at the lower end of the published quinone RFBs (Figure 2). Furthermore, phoenicin also seemed to be less stable than most other quinones as the capacity decay was found to be 2.85%/day. However, one of the most promising prospects of using fungi to biosynthesize quinone electrolytes is the economic costs involved in the production. The phoenicin used to generate the first fungal battery was isolated from the filamentous fungus Penicillium atrosanguineum, which was able to produce 1.24 g/L/week in a liquid medium.⁵⁷ This has later been surpassed in other Penicillium species, where yields of 5 g/L/week were reached in Penicillium phoeniceum.⁵⁹ This potent quinone production is not unique in the fungal kingdom as an even higher yield of 6.83 g/L/ week of bikaverin was obtained in Fusarium fujikuroi.⁶⁰ These studies were based on wild-type strains, which require synthetic growth media to reach high yields. To obtain even higher yields, the quinone biosynthetic gene clusters can be overexpressed in the native host or moved

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FIGURE 4 Overview of selected fungal quinones and their simulated redox potential vs the standard hydrogen electrode (E⁰).⁵⁰

to a fungal production strain for heterologous expression.⁶¹⁻⁶⁵ These methods allow bypassing the normal gene regulation using generic constitutive promoters, the use of standardized production and extraction systems as well as cheap culture media.

Genetic engineering also offers the possibility to modify the biosynthetic pathways to create quinones with superior stability and electrochemistry. So far, the approach of metabolic engineering has not been applied to fungal quinones but is widely used in the pharmaceutical industry in drug development.⁶⁶ Computational simulations revealed a huge width in the redox potentials (E^0) of the known fungal quinones, ranging from -1.298 to 1.485 V vs the standard hydrogen electrode (exemplified in Figure 4), which can be partly explained by differences in side chains.⁵⁰ The effect of the side chains has been shown in a study of artificial guinones, which demonstrated that certain side groups (-OH, -NH₂, and -N $[CH_3]_2$) effectively decrease the E^0 , while -COOH, -CHO, -PO₃H₂, -COOCH₃, -SO₃H, -CF₃, -CN, and -NO₂ is useful in increasing the E^{0.67} Thus, adding some of these side chains to fungal quinones through metabolic engineering can be used in the future to enhance the electrochemical properties of fungal quinones. Here, hydroxylations and methylations are obvious candidates as the required enzymes are widespread among the fungal biosynthetic pathways.

Based on these observations we are hypothesizing that an economically feasible production of quinone electrolytes can be obtained by using genetic engineering to create the optimal cellular metabolic fluxes to produce optimal quinone electrolytes. When this has been achieved, fungal quinones offer a renewable alternative to petrochemical-derived quinones in flow batteries.⁶⁸ Based on the current situation we are therefore in early stages of this process and much more research and development is needed to reach the goal of a commercially available fungal RFB.

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CONFLICT OF INTEREST

The authors declare no conflict of interest.

DATA AVAILABILITY STATEMENT

Data sharing not applicable to this article as no datasets were generated or analysed during the current study.

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