



Efficiency improvement of an all-vanadium redox flow battery by harvesting low-grade heat



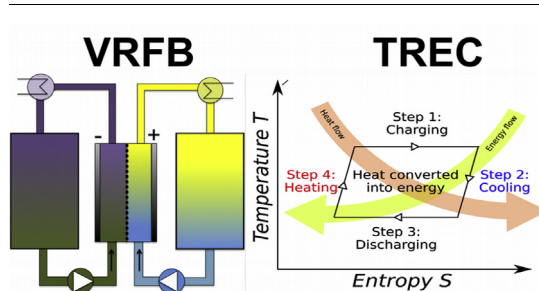
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HIGHLIGHTS

- A *Thermally Regenerative Electrochemical Cycle* combined with a *V-Redox Flow Battery*.
- Operation between 20 and 60 °C.
- Comparison of a *Commercial* and a *Mixed-Acid Electrolyte*.
- Efficiency gain of 9% points (1.3 Wh L⁻¹) for the *Commercial Electrolyte*.
- Efficiency gain of 5% points (0.8 Wh L⁻¹) for the *Mixed-Acid Electrolyte*.

GRAPHICAL ABSTRACT



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ABSTRACT

Redox flow batteries (RFBs) are rugged systems, which can withstand several thousand cycles and last many years. However, they suffer from low energy density, low power density, and low efficiency. Integrating a Thermally Regenerative Electrochemical Cycle (TREC) into the RFB, it is possible to mitigate some of these drawbacks. The TREC takes advantage of the temperature dependence of the cell voltage to convert heat directly into electrical energy. Here, the performance increase of a TREC-RFB is investigated using two kinds of all-vanadium electrolyte chemistries: one containing a typical concentration of sulfuric acid and one containing a large excess of hydrochloric acid. The results show that the energy density of the system was increased by 1.3Wh L⁻¹ and 0.8Wh L⁻¹, respectively and the overall energy efficiency also increased by 9 and 5 percentage points, respectively. The integration of the heat exchangers necessary to change the battery temperature is readily facilitated by the design of the redox flow battery, which already utilizes fluid circulation loops.

1. Introduction

Nowadays, traditional resources such as nuclear or fossil fuels are the most exploited energy resources for electricity generation, but their environmental impact exposes the world to serious concerns. Fortunately, the quest for renewable energy resources has become a priority to satisfy the continuous increase of energy demand [1,2]. However, the intermittent availability of renewable energy resources, such as wind or solar, seriously complicates their integration into large-scale electrical grids. Currently, the intermittence of energy demand is

offset by allocating traditional generating capacity as 'operating reserves'. These reserves can respond quickly to a peak in energy demand. Nevertheless, they are both expensive and rather inefficient [2,3]. As an alternative, batteries are being strongly investigated to facilitate the integration of renewable energy sources, by storing energy during periods of high supply, and supplying energy during periods of shortage. To-date, lithium-ion batteries (Li-ion), sodium-sulfur batteries (Na-S), and redox flow batteries (RFB) have received the greatest consideration for large scale energy storage due to their favorable characteristics (Table 1) [2,4].

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Table 1
 Technical characteristics for Lithium-ion (Li-ion), Sodium-Sulfur (Na-S) and Redox Flow Battery (RFB) [4–6].

	E density [Wh kg ⁻¹]	Cycle life-time	Operating T [°C]
Li-ion	100 to 200	600 to 1200	-10 to 40
Na-S	120 to 150	2500 to 4500	300 to 350
RFB	10 to 50	> 10000	10 to 50

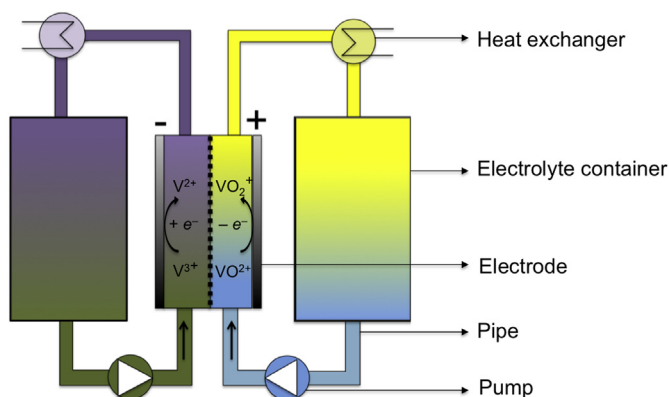
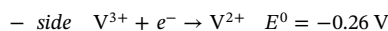
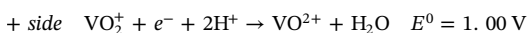


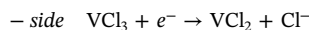
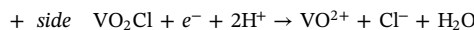
Fig. 1. Schematic of an all-vanadium redox flow battery (VRFB) during charging with two heat exchangers integrated in the piping system.

Redox flow batteries are distinct from Li-ion and Na-S batteries in that the former have a system architecture that includes tanks, pumps, a central reactor, etc., which is analogous to many industrial chemical processes (Fig. 1). Long cycle lifetime is facilitated by the fact that the electrodes are inert spectators of the reaction, and the soluble redox species cannot be consumed. Despite these advantages, RFBs have difficulty competing with other batteries due to their low energy density.

Among the various RFB chemistries, the all-vanadium redox flow battery (VRFB) has received considerable attention (Fig. 1) [3,7]. First studied by Skyllas-Kazacos and co-workers in the 1970s, the VRFB is unaffected by cross-contamination of the redox species because it utilizes the same species on both the negative (V^{III}/V^{II}) and the positive side (V^V/V^{IV}) [1]. Commercial VRFB electrolyte typically contains ca. 1.6 M of the vanadium redox species in ca. 2 M sulfuric acid [8]. In acidic conditions, the following reactions take place at the electrode interface: [8]



Kausar et al. observed that adding ca. 1% phosphoric acid in the electrolyte solution increases the thermal stability of V^V [9]. Similarly, Pacific Northwest National Laboratory (PNNL) added a large excess of hydrochloric acid to the electrolyte yielding better vanadium solubility (2.5M) and a wider thermal range (-10 °C – +50 °C). In the presence of chloride, the resulting reactions taking place in the battery become: [10,11]



Although the standard potential for the vanadium redox couples in the mixed-acid electrolyte was not specified, it was noticed that the reversibility was comparable to the conventional electrolyte, and a small shift in potential for both redox couples was observed providing a larger cell voltage.

Apart from optimising the chemistry of the battery, one possibility to improve the performance is to focus on the thermodynamics of the battery. Taking advantage of the thermogalvanic effect, which is the temperature dependence of the cell voltage, it is possible to convert heat directly into electrical energy [12–14].

Usually a battery is charged and discharged at a constant temperature. However, the thermodynamic potential of the electrode reactions is dependent on the operating temperature. Based on this principle, a thermodynamic cycle may be designed by charging the battery at a higher or lower temperature than the discharge. In this cycle, traditionally-employed compression and expansion of a gas, e.g. Carnot cycle, is replaced by injection and harvesting of electrical charge (Fig. 2 a). Such a system is called Thermally Regenerative Electrochemical Cycle (TREC) and has been reviewed by Chum and Osteryoung in 1981 [15]. Yet, it was primarily studied for high-temperature heat recovery applications. However, to avoid potential overheating and evaporation, for an aqueous-base battery, low temperature heat is desirable. This is ubiquitously available as waste heat (low-grade heat < 100 °C) [14] from many industrial processes, although it requires large heat exchangers to be employed.

In recent years, several TRECs utilizing low-grade heat recovery were developed. In 2014, a TREC using copper hexacyanoferrate and Cu/Cu²⁺ electrodes for low-grade recovery application was proposed [13]. A similar cycle could be performed without membrane [16] or even without an external voltage input [17]. A TREC with supercapacitor was also demonstrated [18]. Long and Li performed several analyses on TREC systems [19].

A TREC can be readily applied to RFBs. As shown in Fig. 1, heat

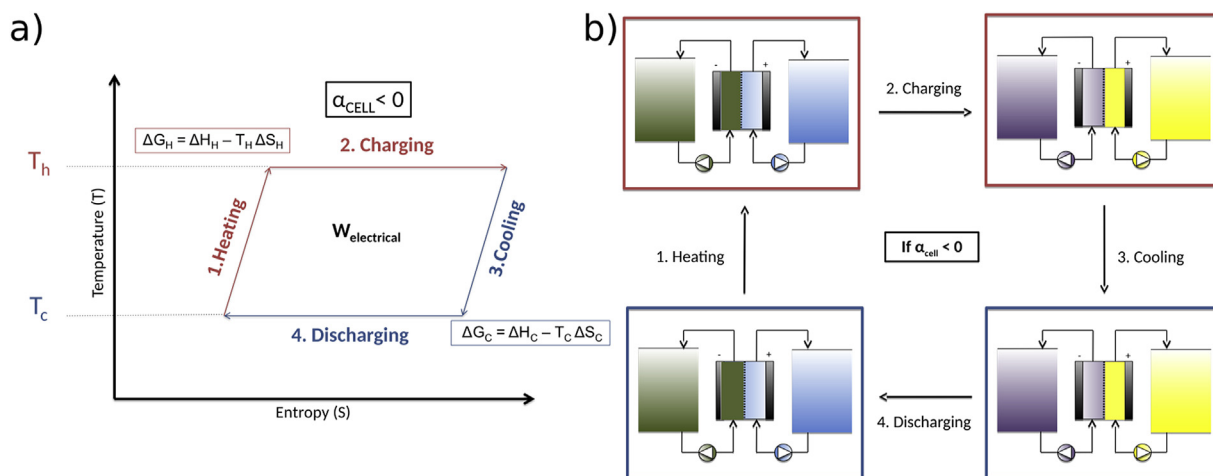


Fig. 2. Schematic of the thermodynamic cycle performed by a TREC (a) Temperature-entropy plot during a cycle performed by a TREC for $\alpha_{\text{cell}} < 0$ and $E_{\text{cell}} < E_{\Delta H}$ [13] (b) Schematic representation of the thermodynamic cycle performed by the TREC-VRFB.

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