

The characteristics and performance of hybrid redox flow batteries with zinc negative electrodes for energy storage

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ABSTRACT

Zinc negative electrodes are well known in primary batteries based on the classical Leclanché cell but a more recent development is the introduction of a number of rechargeable redox flow batteries for pilot and commercial scale using a zinc/zinc ion redox couple, in acid or alkaline electrolytes, or transformation of surface zinc oxides as a reversible electrode. The benefits and limitations of zinc negative electrodes are outlined with examples to discuss their thermodynamic and kinetic characteristics along with their practical aspects. Four main types of redox flow batteries employing zinc electrodes are considered: zinc-bromine, zinc-cerium, zinc-air and zinc-nickel. Problems associated with zinc deposition and dissolution, especially in acid media, are summarized. The main features of each battery are identified and the benefits of a flowing electrolyte are explained. In each case, a summary of their development, the electrode and cell reactions, their potentials, the performance of the positive and negative electrodes, the advantages of a single flow compartment and cell developments for energy storage are included. Remaining challenges are highlighted and possibilities for future advances in redox flow batteries are suggested.

1. Introduction

1.1. Energy storage and sustainability

The need for renewable energy arises from the realisation that fossil fuel supplies are being rapidly depleted with adverse environmental impacts, coupled with the ever-increasing growth of energy consumption due to the growing global population and the demand for higher standards of living [1,2]. Renewable energy can be produced through several means: solar, wind, hydro, tidal, geothermal and biomass. The integration of these carbon-free technologies into the electric grid contributes towards a safe, reliable, flexible, affordable and sustainable energy supply which is essential for the continuous development of a modern society [3]. Several institutions have recognised the importance

of this such as the European Commission who initiated a Renewable Energy Directive for 2020 [4] proposing that 20% of energy consumption in the EU should be derived from renewable sources. This has now been further supplemented by a proposal published in 2016 for the share of energy consumption by renewables to be increased to 27% by 2030 [5]. Likewise, China's National Energy Administration (NEA) has set an ambitious target for 50% of energy consumption in China to be generated by renewable sources by 2020 [6].

In spite of this, the generation of energy from renewable sources is problematic as energy produced is usually in the form of secondary energy [7] making matching supply to demand a complex issue. An Energy Storage System (ESS) is recommended as a viable solution to the intermittency of renewable energy generation [8–10] and seasonal consumer demand as it absorbs the surplus generation capacity,

Abbreviations: 2-D, bi-dimensional; 3-D, three-dimensional; 3NCP, N-benzyl-3-carboxypyridinium chloride; BOD, bilirubin oxidase; CAD, computer aided design; CB, carbon black; CDF, computational fluid dynamics; CNT, carbon nanotube; CTAB, cetyltrimethylammonium bromide; DMAQ, dimethyl-2,6-dihydroxyanthraquinone; DPE, discrete particle electrolyser; ESS, energy storage system; FrTSCN, furfuraldehydethiosemicarbazone; GDE, gas diffusion electrode; HDPE, high density polyethylene; NASICON, sodium super ionic conductor; MEA, membrane electrode assembly; MEM, N-methyl-N-ethylmorpholinium bromide; MEP, N-methyl-N-ethyl-pyrrolidinium bromide; MSA, methanesulfonic acid; MWCNT, multi-walled carbon nanotube; PA, polyamine; Pani, polyaniline; PGE, polymer gel electrolyte; ppi, pores per inch; Pt/Ti, platinised titanium; PTFE, polytetrafluoroethylene; PVA, polyvinyl alcohol; PVE, polyvinyl ester; PVDF, polyvinylidene fluoride; PZ, piperazine; R&D, research and development; RFB, redox flow battery; RTIL, room temperature ionic liquid; RVC, reticulated vitreous carbon; SEM, scanning electron microscopy; SCE, saturated calomel electrode; SHE, standard hydrogen electrode; SOC, state of charge; SWCNT, single-walled carbon nanotube; TBAB, tetrabutylammonium bromide; TEMPO, 2,2,6,6-tetramethylpiperidinyloxy; UPS, uninterruptable power supply; XRD, x-ray diffraction

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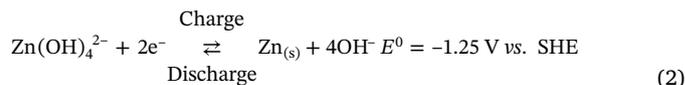
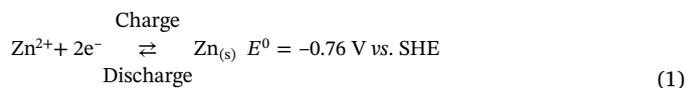
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smooths residual loads and provides flexibility for energy balance [11]. There are several ESS technologies which are near to, or currently, commercially available such as pumped-hydro, compressed air, flywheel and rechargeable battery systems. These systems have been reviewed [12–17] in detail based on their technical, economic and environmental features to give a good indication of their relevance to different applications.

Typically, the generation of energy from renewable sources is carried out on a much smaller scale than conventional power plants, commonly in the range of kilowatts to megawatts, with various levels of applications ranging from small off-grid communities to grid-scale storage [18]. These requirements are suitably met by redox flow batteries (RFBs), first developed by NASA in the 1970s [19] as an electrical energy storage technology. RFBs have proven to be able to support the transmission and distribution of renewable energy efficiently within the grid due to their high cycling efficiency, reasonable nominal duration, reliable long lifespan and flexibility in terms of geographical location [20]. In addition to renewable energy applications, RFBs are also suited to frequency response and UPS services, due to their fast response time, in the order of seconds [7,17]. Other advantages of RFBs, which include their ability to operate at ambient temperature and pressure, absence of harmful emissions, safety in comparison to lithium-ion batteries and greater efficiency in comparison to electrolysis cells, make them ideal medium to large-scale energy storage solutions [21].

1.2. Zinc-based redox flow batteries

Recently, the Zn(II)/Zn redox couple has received considerable interest as the negative electrode reaction in several RFBs for renewable energy storage. Some of the advantages of using this redox couple include: a negative standard electrode, the high solubility of Zn(II) ions, fast kinetics as well as the low cost, abundance and recyclability of zinc compounds. Depending on the chemistry of the RFB, the electrodeposition and dissolution reaction can take place either in acid or in alkaline media [22], respectively:



The standard potentials of common positive electrode reactions and parasitic reactions in zinc RFBs are given in Table 1. It can be seen that numerous RFB chemistries are possible with acid and alkaline electrolytes, as shown in Table 2. In general, high cell potentials are achieved by these systems compared to other proposed RFBs. Thermodynamically, the main competing reaction is H₂ evolution at the Zn electrode, and thus high coulombic efficiencies are only attainable by inhibiting this process. H₂ evolution must also be avoided during open-circuit, as it can result in self-discharge via proton reduction.

As shown in Fig. 1, several RFBs have taken advantage of these properties, exhibiting the highest cell potentials among aqueous systems, which are at least 400 mV higher than the all vanadium cell in most cases. Yet, Zn-based RFBs compromise with relative low charging efficiency, difficult uniform zinc electrodeposition and some degree of self-discharge [28]. The controlled electrodeposition of zinc is critical to produce compact, dendrite-free deposits, uniformly coating the negative side of the bipolar electrodes, in order to prevent short-circuiting. Similarly, uniform and efficient dissolution (oxidation) of the metallic deposit is necessary to avoid its accumulation over repeated battery cycling.

A timeline of developments in rechargeable zinc batteries is presented in Fig. 2. These cells have a long history and, in some cases, have achieved high reliability, e.g., classical Leclanché's zinc/carbon cells [42,43]. Nickel/zinc and zinc/air batteries are also well-known. In the field of RFBs, the zinc-bromine system is the most researched and commercialised, having almost 40 years of development [44]. In contrast, zinc-air and zinc-cerium RFBs continue under investigation, while zinc-nickel RFB has the potential to be developed into economic, undivided cells. Recent years have witnessed a diversity of new chemistries, such as zinc-iron or zinc-polyiodide.

Table 1
Positive electrode reactions found in zinc-based RFBs and their standard redox potentials.

Electrode reaction	Standard electrode potential, E° vs. SHE/V
$\text{Ce}^{4+} + \text{e}^{-} \rightleftharpoons \text{Ce}^{3+}$	+1.74
$\text{O}_2 + 4\text{H}^{+} + 4\text{e}^{-} \rightleftharpoons 2\text{H}_2\text{O}$	+1.23
$\text{Br}_2 + 2\text{e}^{-} \rightleftharpoons 2\text{Br}^{-}$	+1.06
$\text{VO}_2^{+} + 2\text{H}^{+} + \text{e}^{-} \rightleftharpoons \text{VO}^{2+} + \text{H}_2\text{O}$	+1.00
$\text{Fe}^{3+} + \text{e}^{-} \rightleftharpoons \text{Fe}^{2+}$	+0.77
$\text{I}_3^{-} + 2\text{e}^{-} \rightleftharpoons 3\text{I}^{-}$	+0.54
$2\text{NiO}(\text{OH}) + 2\text{H}_2\text{O} + 2\text{e}^{-} \rightleftharpoons 2\text{Ni}(\text{OH})_2 + 2\text{OH}^{-}$	+0.49
$\text{O}_2 + 2\text{H}_2\text{O} + 4\text{e}^{-} \rightleftharpoons 4\text{OH}^{-}$	+0.40
$\text{Fe}(\text{CN})_6^{3-} + \text{e}^{-} \rightleftharpoons \text{Fe}(\text{CN})_6^{4-}$	+0.36
$2\text{H}^{+} + 2\text{e}^{-} \rightleftharpoons \text{H}_2$	0.00

Table 2
Selected cell reactions relevant to proposed zinc-based RFBs together with their standard cell potential.

Cell type	Discharge cell reaction	Standard cell potential, $E^{\circ}_{\text{cell}}/\text{V}$	Ref.
Zn-Ce	$\text{Zn} + 2\text{Ce}^{4+} \rightleftharpoons \text{Zn}^{2+} + 2\text{Ce}^{3+}$	2.48	[23]
Zn-Cl ₂	$\text{Zn} + \text{Cl}_2 \rightleftharpoons \text{Zn}^{2+} + 2\text{Cl}^{-}$	2.12	[24]
Zn-Br ₂	$\text{Zn} + \text{Br}_2 \rightleftharpoons \text{Zn}^{2+} + 2\text{Br}^{-}$	1.82	[25]
Zn-V	$\text{Zn} + 2\text{VO}_2^{+} + 4\text{H}^{+} \rightleftharpoons \text{Zn}^{2+} + 2\text{VO}^{2+} + 2\text{H}_2\text{O}$	1.76	[26]
Zn-Ni	$\text{Zn} + 2\text{NiO}(\text{OH}) + 2\text{H}_2\text{O} + 2\text{OH}^{-} \rightleftharpoons 2\text{Ni}(\text{OH})_2 + \text{Zn}(\text{OH})_4^{2-}$	1.71	[27]
Zn-air	$2\text{Zn} + \text{O}_2 \rightleftharpoons 2\text{ZnO}$	1.65	[28,54]
Zn-ferricyanide	$\text{Zn} + 2\text{Fe}(\text{CN})_6^{3-} + 4\text{OH}^{-} \rightleftharpoons 2\text{Fe}(\text{CN})_6^{4-} + \text{Zn}(\text{OH})_4^{2-}$	1.61	[29]
Zn-Fe	$\text{Zn} + 2\text{Fe}^{3+} \rightleftharpoons \text{Zn}^{2+} + 2\text{Fe}^{2+}$	1.53	[24,30]
Zn-polyiodide	$\text{Zn} + \text{I}_3^{-} \rightleftharpoons \text{Zn}^{2+} + 3\text{I}^{-}$	1.30	[31]

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