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Development and testing of an economic grid-scale flow-assisted zinc/nickel-hydroxide alkaline battery

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HIGHLIGHTS

- We present a new design of nickel zinc battery at bench scale and grid scale.
- We present cycle testing, failure analysis and gas sealing of this new battery.
- Zinc particulates and nickel surface passivation are the primary failure mechanisms.
- Cycle life of over 3000 is achieved and cost estimates show low cost, $407 per kWh.

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ABSTRACT

An economic design for an alkaline zinc-anode flow-assisted battery without membrane separators was tested at grid-scale of 25 kWh with a string of thirty 833 Wh cells in series, and also at bench scale with individual 28 Wh cells. The bench-scale tests allowed optimization of parameters such as electrolyte flow, choice of hardware material, electrolyte composition, and charge/discharge protocol. The best-performing bench scale cell cycled for over 3300 cycles with energy efficiency above 80%, and was selected as the design basis for scale-up to the 25 kWh battery string. Testing of the grid-scale string demonstrated 1000+ cycles with round trip energy efficiency above 80%. Two challenges observed at the bench scale were overcome for successful scale-up, namely a) passivation of the anode surface, which occurred when the anode experienced voltages 100 mV above zinc's rest voltage, and b) zinc particulates that jammed the gap between the electrodes and caused cathode degradation and passivation of the anode surface. Best practices to overcome these challenges and achieve long cycle life are presented.
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1. Introduction

Electrical energy storage at MWh scale is under widespread development as a means to improve grid stability in the increasingly sophisticated electric power market. The leading motivation for energy storage is growth of solar and wind power, which at grid-penetration above ~20% requires energy storage to smooth out variability [1,2]. Other applications with significant demand for energy storage are nighttime load shifting, demand-charge reduction and uninterruptible power supply applications. Market analysis typically suggests target costs for the grid-scale battery system to be less than $500 per kWh, with cycle life above 2000 and discharge capability that allows total capacity to be delivered in less than two hours [3–6]. These performance numbers mean new battery technology will have to be developed. Existing lead-acid batteries are unlikely to be suitable because they cannot discharge their capacity in less than three hours without significant loss of capacity or cycle life. Widespread manufacturing and use of lead is also undesirable due to environmental concerns. Existing nickel–zinc batteries are limited by the early failure of the pasted zinc anode, after ~700 cycles [15], which causes the cost per lifetime kWh-delivered to be too high. Fire safety is also likely to play a

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role. In residential or urban locations the fire risks of the batteries are required to be very low, and relatively high fire risk exists for lithium ion, molten-metal, or sodium sulfur technologies. To work toward meeting the market requirements, this paper reports further development of flow-assisted alkaline batteries with zinc anodes and sintered-nickel nickel-hydroxide cathodes [7,8], based on the reactions

$$\text{cathode}: \quad 2NiOOH + 2H_2O + 2e^- \Leftrightarrow 2Ni(OH)_2 + 2OH^- \tag{1}$$

$$\text{anode}: \quad Zn + 4OH^- \Leftrightarrow Zn(OH)_4^{2-} + 2e^- \tag{2}$$

in 9 M KOH electrolyte, which meet the performance goals and have negligible risk from fire or chemical hazard. Our "flow-assist" battery is not the same as a flow battery, but simply means that the electrolyte is circulated inside each cell, through flow gaps between the electrodes, to improve the electrodeposition and dissolution of zinc during long-term cycling.

We have conducted and published results of previous experiments [7,8] with the same basic technology and demonstrated cycle life up to 1500 in 3.7 Ah cells. These results indicated that the coulombic efficiency and discharge capacity were improved if electrolyte flow velocity was increased above 10 cm s^{-1}. The pumping costs are significant at these flow-assist velocities therefore we choose flow rates of 0.5–1 cm s^{-1} which are shown in Subsection 3.3 to require only 4% of the energy stored in the battery cell.

2. Experimental

2.1. Bench-scale cell testing

Bench scale tests were conducted with 19 Ah (31 Wh) cells of specifications shown in Table 1. The goal of the bench scale tests was to optimize design of the larger grid-scale cells that would comprise the 25 kWh battery. Fig. 1 shows CAD drawings of a bench scale cell. A polysulfone box of 1.5 L volume (outside dimensions) contained the electrolyte and electrodes. The electrode plates stood upright, parallel to each other, with cathode plates separated from anode plates by a 3 mm gap of upward-flowing electrolyte created with 3 mm diameter PMMA dowel rods spaced apart by 15 mm. The electrode plates were sealed inside a PMMA "chimney" box (i.e. with an open top) that was immersed in the electrolyte. The two tube ports seen on the top lid allowed electrolyte to be pumped into the bottom of this chimney box, causing the electrolyte to flow upward between the electrode plates, as shown with the arrows of the right side drawing of the figure. At the top of these channels the flow was restricted by a narrow gap, which homogenized the flow of electrolyte to be the same velocity in each channel. The electrolyte then spilled out the top of the chimney and then circulated through the pump again. Table 1 specifies the baseline design of these bench-scale cells (19 Ah, 28 Wh). The sintered-nickel cathodes were obtained from Jiangsu Highstar Battery Company. The nickel-plated anodes were obtained from Orbel Corporation. All other chemicals were obtained from Fischer Scientific or Sigma Aldrich, and all other hardware from McMaster Carr. Nuts, bolts and hardware were electroless nickel plated.

Table 1
Bench scale cell design (baseline design).

Design parameter	Value
Electrolyte volume	0.95 L
Electrode size	10 × 9 cm, with some regions masked
Number of anodes	9
Number of cathodes	8
Gap between electrodes	3 mm
Cathode material	0.66 mm Highstar sintered nickel cathode
Cathode discharge capacity	0.03 Ah cm^{-2}, total for cell is 19 Ah
Anode material	0.14 mm copper sheet coated with 5 μm of nickel
Anode electro-deposition capacity	60 g L^{-1} of ZnO in 0.95 L equates to 38 Ah
Zincate concentration in electrolyte	60 g ZnO L^{-1}
Potassium hydroxide concentration in electrolyte	45% w/w
Energy density	20.9 Wh L^{-1}
Specific Energy	17.3 Wh kg^{-1}
Flow velocity upward between electrode plates	0.5 cm s^{-1}
Material of bolts, tabs, hardware	Nickel coated steel or nickel coated copper

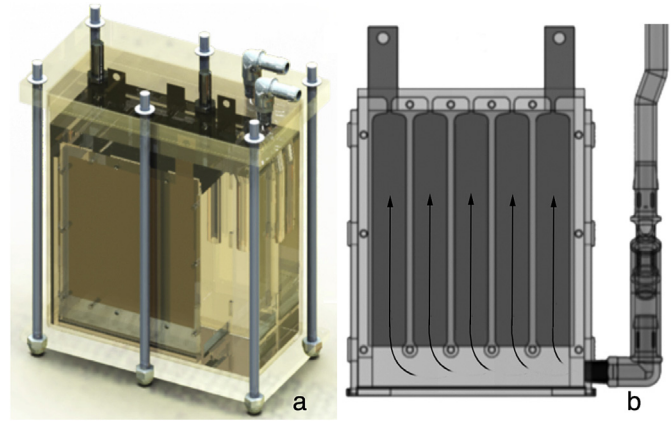

Fig. 1. a) The bench-scale cell is shown. b) A cross-section of the "chimney" box that was immersed in the electrolyte is shown, where the electrodes are dark grey and PMMA dowel rods are light grey.

The open-circuit voltage of the bench and grid scale cells ranged from 1.50 V at low state-of-charge to 1.80 V at high state-of-charge. This change in cell voltage was due to the nickel hydroxide cathode [9]. The voltage of the zinc anode remained always within 20 mV of zinc's reference voltage in the same electrolyte. Cycling was performed at 100% depth of discharge with an Arbin Instruments BT-2000 battery tester. The cycling protocol charged the cells at a constant current rate of "full capacity in two hours" (9.5 A). The charging switched to constant voltage at 1.93 V, to avoid excessive gassing of oxygen. Discharge of the cells was also performed at constant current "full capacity in two hours" (−9.5 A), ending when the cell voltage dropped below 1.2 V. These charge/discharge rates allowed roughly five cycles to be tested per day. The upward flow velocity of electrolyte between the electrode plates was ensured to be 0.5 cm s^{-1}. As described in Ref. [8] the cells were put through an "anode cleaning" every 10th cycle to remove excess zinc off the anodes. The anode cleaning procedure discharged the cells at constant current "full capacity in ten hours" (−2.0 A) until the cell voltage reached −0.1 V, after every tenth cycle. This cleaning discharge current of −2.0 A was found to be too high for optimal performance, but a full description of this is left for Subsection 3.1. A list of our bench scale tests and results is given in Table 2 in Section 3.1.

2.2. Gas sealing the cell, and maintaining proper electrode state-of-charge

Metallic zinc is well known to experience a corrosion reaction with the electrolyte that slowly dissolves the zinc and produces gaseous bubbles of hydrogen [10,12,13]. The anode's nickel surface

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