Li/Li_2S_x Liquid Cathode Cell

Alexander Gorkovenko, Eugene Tritenichenko,
Charles Skaggs, and Vince Puglisi
Moltech Corporation, Tucson AZ

Abstract

The high specific capacities of sulfur [1600 mAH/g] and lithium [3800 mAH/g], with the combined specific energy exceeding 2500 WH/kg, is a driving force behind the development of a rechargeable Lithium/Sulfur battery. Due to performance considerations, the majority of attempts to create a practical battery rely on liquid cathode systems. In this paper, we present the first true secondary Li/Li_2S_x liquid cathode cell, Table 1.

The advantages of Li/S systems are well known: high specific energy, good rate capability, low temperature performance, and "built in" overcharge protection. Typically, the biggest shortfall exhibited with these systems is a difficulty to sustain long cycle life. There are two reasons for this shortfall. First it is not trivial to stabilize the sulfur electrode because the Li_2S_x reduction/oxidation is a complex multi-step process which includes phase separation of several intermediate products. Second, the electrolyte, or more correctly, the liquid cathode, is a variable. That is, the composition and concentration of the liquid cathode is a function of the sulfur electrode's state of charge at any given time. The lithium electrode's cycling efficiency, at the same time, is controlled by the nature of the solid electrolyte interface (SEI) film formed *in situ* on it's surface and/or by surface treatment prior to assembly. The issue is that the changing liquid cathode environment makes it difficult to design a well functioning, stable SEI film.

Typical cycle life of Moltech's Li/Li_2S_x liquid cathode cell had been on the order of 150-200 cycles at 80 % of rated capacity. This equates to a Figure of Merit (FOM) of 30-35. The capacity loss was approximately 0.1-0.13 %/cycle during the cell's life. Most applications require a minimum of 300 cycles, with 500 cycles being desirable. Recent dramatic advances in the chemistry of the cell, while leaving the cell's mechanical design unchanged have resulted in an improved version of the Li/Li_2S_x battery. This cell type delivered an average of 378 cycles at 80 % of rated capacity. This equates to a FOM of 62. The capacity loss was approximately 0.02%/cycle during the cell's mid-life. The specific energy of this cell design was 145 WH/kg. The cell was packaged in an non-optimized aluminized PET soft pack. The cell was charged at 200 mA and discharged at 350 mA to 1.8V.

The cycle life can be considered as having four stages as illustrated in Fig. 1 and described below:

I. Dissolution of sulfur, formation of liquid Li_2S_x cathode after first discharge. Cell discharge capacity is rated on 5th cycle, Table 1, and Fig. 2.
II. Stabilization of sulfur electrode performance: first 70-100 cycles of cell cycle life. Cell capacity decay rate at this stage is 0.11 %/cycle.
III. Lithium and sulfur electrode performance is stable, next 250 cycles. Cell capacity decay rate at this stage is 0.02 %/cycle, Fig. 3.
IV. Deterioration of lithium electrode in the last 50 cycles. Cell capacity drops to 80% of Q_5.

All previously described Li/S cells were devoid of the stable stage in their life span. Only stages I, II, and IV were present and the sulfur electrode formation stage was superimposed onto the lithium electrode's capacity decay.

Table 1. Cell build parameters (15 cells):
Mean value/Standard Deviation

Capacity 5th cycle, mAH	Spec. Energy, WH/kg	Cell Weight, g	FOM @80% Q_5	Cycles @80% Q_5
812/6	145/2	11.6/0.1	62/3	378/21

Fig. 1 Four stages of Li/Li_2S_x cell cycle life

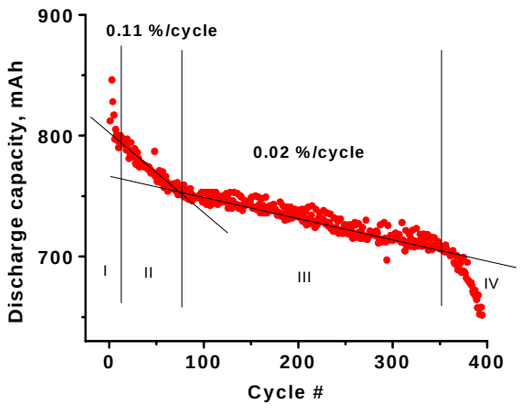


Fig. 2 Voltage/time discharge curves, 1st and 5th cycle

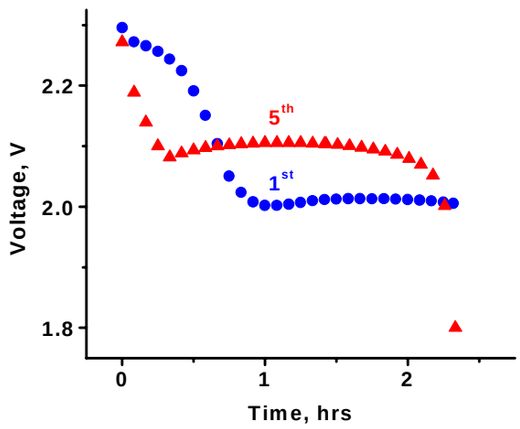
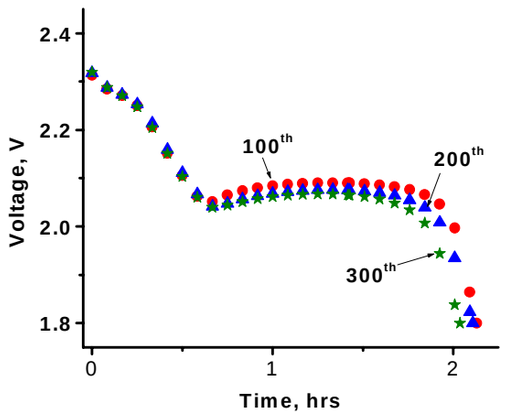


Fig. 3 Voltage/time discharge curves, 100th, 200th and 300th cycle



Acknowledgement: The authors wish to thank Mr. Zhe-Sheng Xu for his contributions to this effort.