

# Lithium Sulfur Pouch Cells with Improved Energy Density for Portable Power Applications

**Ernest Ndzebet, Mario Destephen, Logan Wittmaier and Anulekha Haridas**

Research and Development  
EaglePicher Technologies  
1215 West C Street, Joplin, MO 64801  
[ernest.ndzebet@eaglepicher.com](mailto:ernest.ndzebet@eaglepicher.com)

**Abstract:** *The Lithium-Sulfur (LiS) battery has been considered one of the most promising rechargeable power sources because of its high theoretical energy density, low cost, and environmentally friendly elemental Sulfur cathode material. However, LiS batteries still suffer from historical limitations that include short cycle life, poor shelf life, and low energy density and rate capability. EaglePicher has developed an electrolyte formulation, high loading cathode, and proprietary Nanoclay separator to address these limitations, demonstrating improved performance in a small 2 Ah pouch cell format. A nominal specific energy greater than 400 Wh/kg at room temperature is projected in a larger 10 Ah pouch cell format. Moreover, early prototypes of the LiS pouch cells passed preliminary safety tests and successfully operated in a wide temperature range. These results prompted EaglePicher to build 28 Volt prototype batteries with an internally developed battery management system (BMS).*

**Keywords:** Lithium-Sulfur; Nanoclay separators; shelf life; cycle life; intermittent discharge performance

## Introduction

There has been increasing demand for secondary batteries with lighter weight and higher capacity for portable power applications. Lithium-Sulfur is a promising chemistry which offers a high theoretical capacity of 1672 mAh/g and theoretical specific energy of 2600 Wh/kg [1]. In addition, Sulfur is cheap and environmentally safe, which makes Lithium-Sulfur an attractive chemistry for next generation batteries beyond Li-ion. There are some challenges associated with this chemistry, however. Sulfur ( $5 \times 10^{-30}$  S/cm) and its final discharge product, Lithium sulfide, are both insulators, requiring a high percentage of Carbon to achieve sufficient cathode conductivity and rate capability [2]. The high carbon content in the cathode can result in LiS cell will low practical energy density.

The discharge of a Lithium-Sulfur battery proceeds in multiple steps. In the first plateau, Sulfur is converted to polysulfides ( $\text{Li}_2\text{S}_n$ ,  $n$  varies from 3 to 8). In the second plateau these polysulfides are reduced to solids,  $\text{Li}_2\text{S}_2$  and finally to  $\text{Li}_2\text{S}$ . The polysulfides ( $\text{Li}_2\text{S}_n$ ,  $3 < n < 8$ ) are soluble in electrolyte and can be deposited as  $\text{Li}_2\text{S}$  either on the

anode or on the cathode. When deposited on the cathode, the  $\text{Li}_2\text{S}$  clogs the pores over multiple charge/discharge cycles. In addition, there is also volume change due to the differences in the molar volumes of S and  $\text{Li}_2\text{S}$ , which affects the cathode structure. Both of these effects lead to a decrease in capacity over the course of the cycle life.

During charging, the  $\text{Li}_2\text{S}$  on the cathode side is oxidized to higher-order polysulfides, which can migrate and get reduced to lower-order polysulfides as  $\text{Li}_2\text{S}$  deposits on the anode. Thus, the soluble polysulfides can shuttle between cathode and anode causing extended charging times and low coulombic efficiency in the Lithium-Sulfur chemistry. This causes further capacity fading over multiple charge/discharge cycles [3]. In addition, upon storage, Sulfur can dissolve in the electrolyte as a polysulfide via a self-discharge mechanism [4], leading to poor shelf-life performance.

Different approaches, including the use of highly porous Carbon materials to confine Sulfur and polysulfides in the cathode structure, have been attempted to address these issues [5]. In addition, the use of redox-active interlayers consisting of Sulfur impregnated polar ordered mesoporous silica was found effective as a means to increase cell capacity and to protect the anode [6].

With respect to electrolyte, numerous reports exist regarding the choice of solvents in liquid electrolyte, the use of gel electrolytes, and polymer electrolytes to reduce the diffusion of polysulfides towards the anode [7, 8]. In some cases, anode protection from polysulfide deposition has been achieved through the use of electrolyte additives [8]. Indeed, the additive  $\text{LiNO}_3$  is known to suppress the redox shuttle of the dissolved polysulfides on the Li anode [9]. Although the above-mentioned approaches led to LiS cells with improved rate capability and cycle life, further work is still needed to increase their practical energy density.

This paper details the development of high energy density LiS pouch cells for portable power applications. In this work, a combined approach that includes advanced high loading cathode development, electrolyte formulation, separator development, and in situ anode protection was

used to improve the energy density and shelf-life performance of LiS cells. Preliminary safety evaluations of these cells and their performance in a battery configuration were also evaluated.

## Experimental

EaglePicher has developed a high energy density LiS pouch cell that includes a web-coated high loading Sulfur cathode consisting of proprietary Carbon materials and separator. This was achieved through extensive development efforts over the last several years. The web coating process enables thinner cathodes with improved rate capability, which contributes to improvements in LiS cell performance. The type of Carbon used as a conducting additive and its distribution within the cathode structure, as well as optimized cathode porosity further enhance the cell rate capability. Carbon and Sulfur were processed in a specified ratio to form a Carbon/Sulfur composite cathode. Our cathode processing is a modified version of the process reported elsewhere [3]. The Carbon/Sulfur composite was mixed with the binder and solvent, and the resulting slurry was coated on an aluminum current collector to form the cathode.

The electrochemical performance of the Lithium-Sulfur cell was evaluated in a stack pouch cell configuration using our proprietary nanoclay/polymer separator, which was made by a solution casting method. Cells were activated with our proprietary electrolyte. The cell performance evaluation was conducted at temperatures ranging from  $-20^{\circ}\text{C}$  to  $60^{\circ}\text{C}$  using an Arbin battery cycler. Cycling was between 2.7 V and 1.7 V (vs.  $\text{Li}^+/\text{Li}$ ) at a rate of  $\geq 0.4 \text{ mA}/\text{cm}^2$ . Li-S cells undergo more severe self-discharge than more typical LIBs due to polysulfide shuttling. Because of this, the cell shelf-life evaluation was carried out through accelerated aging at  $45^{\circ}\text{C}$  up to one (1) month. Preliminary safety evaluations including nail penetration, overcharge, external short circuit, and crush tests were carried out at room temperature.

The performance improvements observed at the cell level prompted us to build a 28V prototype battery consisting of 14 stack pouch cells in series with an in-house developed BMS. The battery was cycled at similar rates at room temperature.

## Results & Discussion

Figure 1 shows the performance of LiS cells discharged at C/8 and room temperature. As can be seen, the cells delivered an average capacity of 1.92 Ah when discharged to 1.7V. They delivered an average specific energy of 213 Wh/kg on the first cycle and 175 Wh/kg on the 30<sup>th</sup>, as illustrated in Figure 2. It is expected that, with further cell design optimization including higher Sulfur loadings and lower electrolyte amounts, this chemistry can deliver 250 Wh/kg in this 2 Ah pouch format. In a larger 10 Ah pouch

cell, a projected nominal specific energy of greater than 400 Wh/kg at room temperature can be achieved.

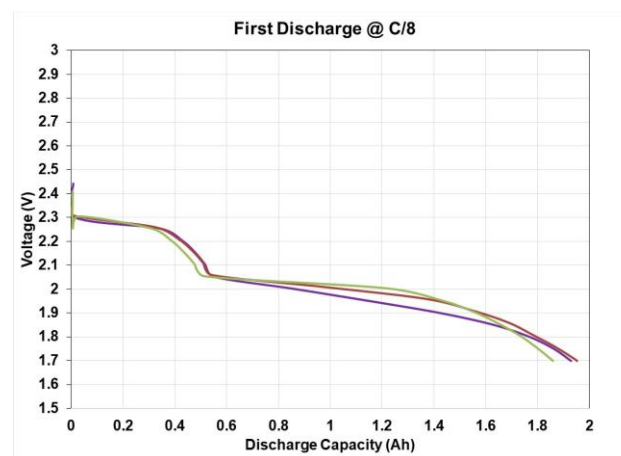


Figure 1. Discharge profile of LiS at room temperature

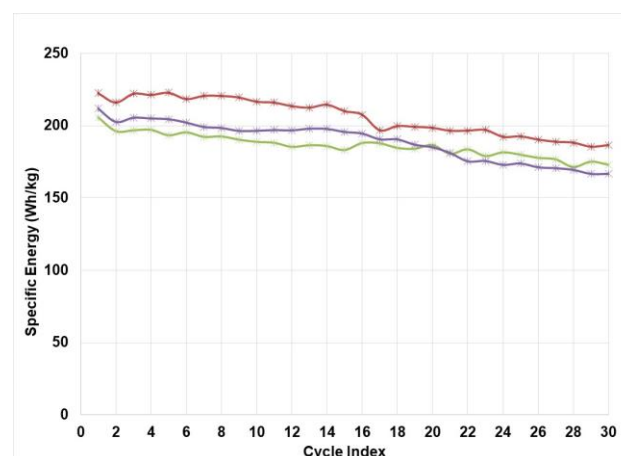


Figure 2. Cycling performance at room temperature

Discharge performance of these cells was also evaluated at temperatures ranging from  $-20^{\circ}\text{C}$  to  $60^{\circ}\text{C}$ , illustrated in Figure 3. As can be seen, cells showed initial capacities higher than 750 mAh/g for most of the investigated temperatures, with higher temperatures yielding higher initial capacities. The lowest performance observed was in cells tested at  $-20^{\circ}\text{C}$ , which can be attributed to an increase in electrolyte viscosity and slow diffusion of polysulfides in the cathode at low temperatures.

The cycling performance of LiS cells at different discharge rates is shown in Figure 4. As can be seen, high capacity and improved cycle life is observed in cells tested at C/8. When tested at rates higher than C/8, cells showed capacities of 700 mAh/g for 10 cycles, but after 30 cycles this is reduced to 500 mAh/g. Further development work focusing on the use of Carbon nanotubes and new binders in the cathode, anode protection, and electrolyte additives is under way to improve cell rate capability and capacity.

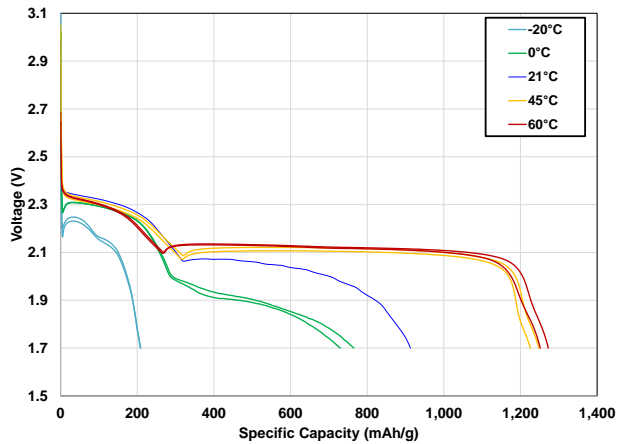


Figure 3. Effect of temperatures on LiS cell performance

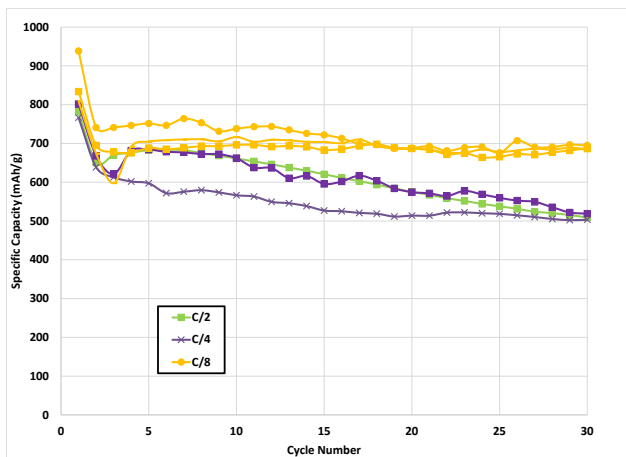


Figure 4. Cycle life performance at different discharge rates

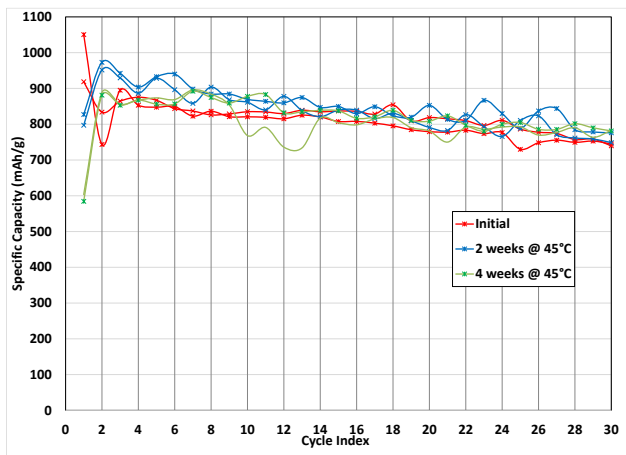


Figure 5. Shelf-life performance of cells after 45C storage

Cells were stored at 45°C for two and four weeks to evaluate their shelf-life performance. Data from fresh cells

is provided for comparison. As can be seen in Figure 5, the initial capacity is lower after two weeks and four weeks of storage compared to that of fresh cells, but the capacity recovered after one discharge/charge cycle. Overall, these cells showed good cycle life with minimum capacity loss over 30 cycles after being stored at 45°C.

Initial safety evaluations were conducted on the LiS pouch cell, and the conditions and results of each test are summarized in Table 1. The cells passed each test without producing fire or smoke. These preliminary results are encouraging, but further testing is still needed for a full safety assessment of this cell design.

Table 1. Preliminary Safety Tests of LiS Pouch Cell

Test	Conditions	Observation	Result
Nail Penetration	-Fresh cells at 100% SOC -After 10 cycles at 100% SOC	No smoke or fire	Passed
Over Charge at 1C rate (2A)	-Fresh cells at 100% SOC -After 10 cycles at 100% SOC	No fire, Cell swelled	Passed
External Short-Circuit (1.5mΩ resistance)	Fresh cells at 100% SOC -After 10 cycles at 100% SOC	No fire, Cell swelled	Passed
Crush Test – 13±0.78 kN @ 1.5 cm/s speed	Fresh cells at 100% SOC -After 10 cycles at 100% SOC	No smoke or fire	Passed

As mentioned earlier, the observed cell performance improvements were achieved through cathode processing and design, along with our proprietary electrolyte formulation and functionalized nanoclay separator. This separator has showed improved performance over commercially available separators, as previously reported [11]. The efficacy of the nanoclay separator is believed to be arising from the ability of its positive functional groups to electrostatically bond with the negatively charged polysulfides.

LiS cells with these improvements were built into a battery in a 14S1P configuration with a BMS that was developed in-house. This yielded a battery pack with nominal voltage and capacity of 28V and 2Ah, respectively. The first prototype battery was cycled at the C/8 rate at room temperature, but was stopped after 5 cycles due to a BMS related error. Figure 6 shows this cycling effort. The first cycle shows low capacity, but it increases after subsequent cycles.

The capacity delivered by the battery is lower than the expected 2Ah due to the nature of the implemented BMS and the configuration of the battery. With cells in series, one poor performing cell will greatly affect the performance of the battery overall. Cell balancing in a series string of cells such as this prototype would require additional development work on a more advanced BMS.

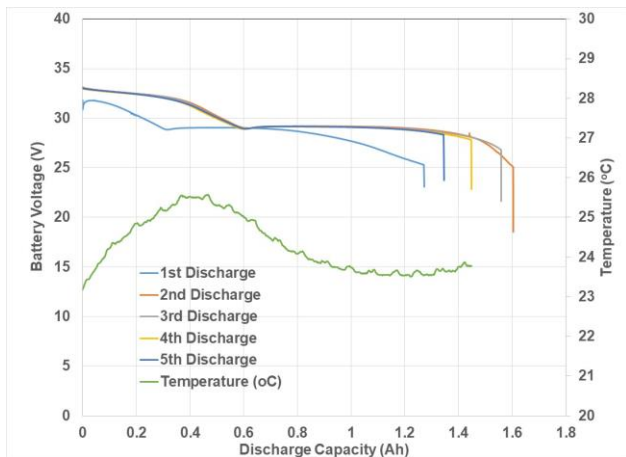


Figure 6. Discharge profile of the prototype battery

A second prototype was made and subjected to more extended cycling, shown in Figure 7. This battery went through more than 30 charge/discharge cycles before testing was stopped. This battery shows lower capacity and higher capacity fade than the individual cells shown earlier, but with a more advanced BMS that can better balance individual cells in the pack, both of these effects can be minimized.

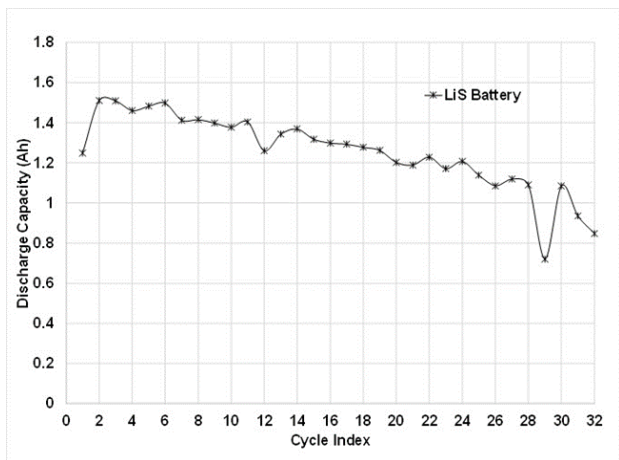


Figure 7. Cycle life of second prototype LiS battery

## Conclusion

EaglePicher has developed 2Ah LiS pouch cells with improved discharge, cycle life, and shelf life performance. This was achieved through use of our proprietary electrolyte formulation, high loading cathode, and proprietary nanoclay separator. A nominal specific energy greater than 400 Wh/kg at room temperature is projected in a larger 10Ah pouch cell format. Moreover, early prototypes of the LiS pouch cells passed preliminary safety tests and successfully operated in a wide temperature range. The performance of these cells was evaluated in 28V, 2Ah prototype batteries with an internally developed

battery management system (BMS) with encouraging results.

## Acknowledgments

This material is released to the public domain in accordance with ITAR 22 CFR 120.11.

## References

1. Ji, X. and L.F. nazar, "Advances in Li-S batteries", *J. Materials. Chem.*, vol. 22, pp. 691-714, 2010.
2. Han, J-S., S-S. Choi, S-H. Park and Y-S. Choi, "Positive electrode for Lithium-Sulfur battery", *US Patent*, no, 7147968B2, Dec. 12, 2006.
3. Ji, X., K.T. Lee and L.F. Nazar, "A highly ordered nanostructured Carbon-Sulfur cathode for Lithium-Sulfur batteries", *Nature Materials*, vol. 9, pp. 500-506, 2009.
4. Yamin, H. and E. Peled, "Electrochemistry of a nonaqueous Lithium/Sulfur cell", *J. Power Sources*, vol. 9, pp. 281-287, 1983.
5. Liang, C., N.J. Dudney and J.Y. Howe, "Sulfur-Carbon nanocomposites and their application as cathode materials in Lithium-Sulfur batteries", for *US Patent*, no. 2011/0052998A1, Mar. 3, 2011.
6. Lee, B.J. et al, "Development of high-energy density non-aqueous lithium sulfur batteries via redox-active interlayer strategy", *Nat Commun* 13, 4629 (2022). <https://doi.org/10.1038/s41467-022-31943-8>.
7. Chu, M.-Y., L.C. De Jonghe, S.J. Visco and B.D. Katz, "Liquid electrolyte Lithium-Sulfur batteries", *US Patent*, no. 6030720, Feb. 29, 2000.
8. Ryou, M.H. et al, "A gel polymer electrolyte based on initiator-free photopolymerization for Lithium secondary batteries", *Electrochim. Acta*, vol. 60, pp. 23-30, 2012.
9. Aurbach, D., E. Pollak, R. Elazari, G. Salitra, C.S. Kelly, J. Affinito, "On the Surface Chemical Aspects of Very High Energy Density Rechargeable Li-Sulfur Batteries", *J. Electrochem. Soc.*, vol. 156, pp. A694-A702, 2009.
10. Zhang, S.S, *Electrochim. Acta* Vol. 70, pp.344-348, 2012
11. Janakiraman, U. et al, *Power Sources Conf.* Vol. 45, 2012