# Development of 3 and 15 Ah Rechargeable Lithium Pouch Cells

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## Abstract

*Commercially available Li-ion batteries using graphite or graphite-silicon blended anodes are currently approaching a cell level specific energy of 350 Wh kg$^{-1}$. This paper will overview the technologies developed for a 3 Ah pouch cell with a specific energy above 400 Wh kg$^{-1}$. Several areas will be highlighted that resulted result in improved cell performance such as increasing the tab size, optimizing the electrolyte type and amount, and introducing excess lithium metal into the anode. The cell delivers ~425 Wh kg$^{-1}$ at low rate and >300 Wh kg$^{-1}$ at 6.6C. Finally, the initial prototype performance of a 15 Ah rechargeable lithium pouch cell with a specific energy >500 Wh kg$^{-1}$ will be highlighted.*

## Keywords

battery; lithium anode; lithium metal; rechargeable; high energy

## Introduction

Lithium metal is an ideal negative electrode material because it is the lightest metal and very electronegative ($-3.04$ V vs the standard hydrogen electrode). These properties lead to a battery that demonstrates extremely high specific energy and energy density. The chemistries that use lithium metal as the anode in figure 1 have the highest practical specific energies at the cell level.

All attempts to commercialize rechargeable lithium metal batteries since they were first proposed in the early 1970s have failed due to poor cycling and safety performance caused by rough Li deposits formed on the negative electrode during cell charging. These deposits are often referred to as mossy lithium and in the worst-case dendrites. Though the exact cause of mossy and dendritic lithium is unknown, it is most likely related to the solid electrolyte interphase (SEI). The SEI is a passivating layer that forms on lithium metal when in contact with nonaqueous electrolyte because most organic solvents are not thermodynamically stable at lithium's low potential. The SEI prevents further reduction of electrolyte components allowing for cell operation. The SEI impacts most areas of battery performance, including power density, cycle efficiency and life, and safety.

The mossy lithium has a very high surface area which leads to additional SEI formation that consumes active lithium. This lowers the cycle coulombic efficiency leading to poor
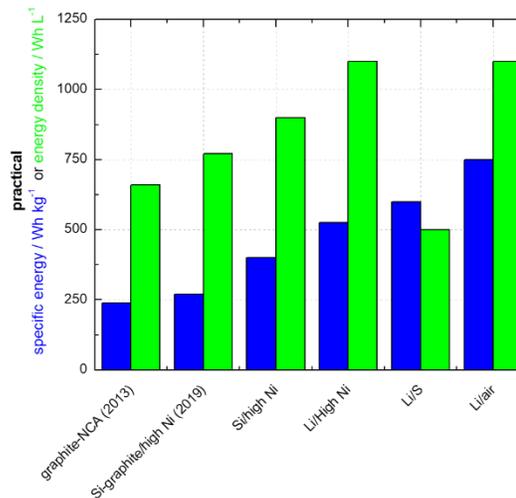


**Figure 1.** Comparison of practical cell-level specific energy and energy density for different lithium battery chemistries.

cycle life. Furthermore, lithium deposits can become electrically isolated from the anode. Electrically isolated lithium deposits are referred to as "dead lithium" and can no longer be discharged leading to lower capacities and poor cycle life. Dendritic lithium deposits have branch-like morphology. The dendrites will continue to grow as the cell cycles and can eventually transverse the separator to the cathode forming a hard short. This will lead to cell thermal runaway and typically explosion. The cells described in this paper fail before any internal short causes an explosion.

Despite the poor cycle life and potential safety hazard, the lithium metal anode is considered the "holy grail" of lithium battery chemistries due to its extremely high specific energy and energy density. One potential application for this chemistry of interest to these proceedings is military unmanned aerial vehicles which often do not fly more than 50 missions. A cycle life of ~100 cycles above 300 Wh kg$^{-1}$ would enable longer times for these missions.
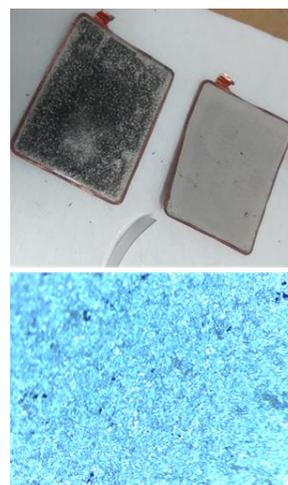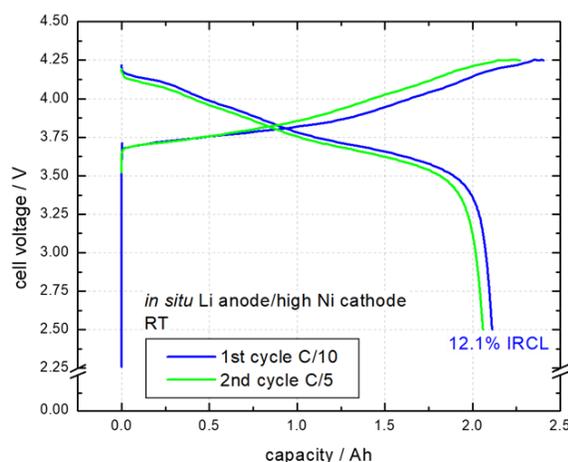
## Anodeless cell design

One possible way to build a cell with a lithium metal anode is to replace a conventional lithium-ion graphite or graphite and silicon composite anode that is web-coated onto copper foil with lithium metal. This design would have active
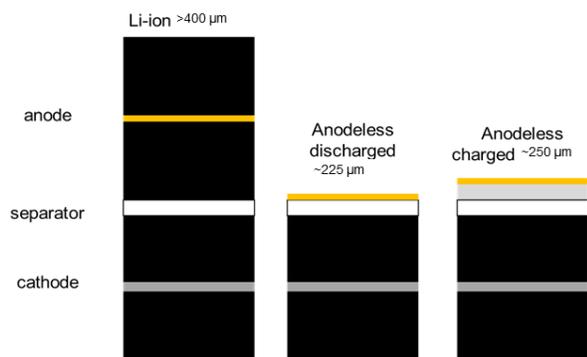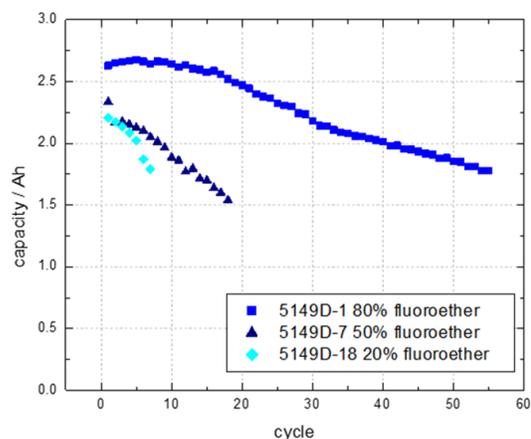
**Figure 2.** Representations of the conventional and anodeless cell designs.



**Figure 4.** Effect of fluorether content on cell cycling performance.

lithium in both the anode and cathode. This excess lithium would lead to better cycle life but only a modest specific energy increase over SOA Li-ion cells. Candidate cathodes that do not contain lithium like $MnO_2$ and metal fluorides (e.g. $CuF_2$, $FeF_3$, etc.) have poor reversibility leading to extremely low cycle life. Others with good reversibility like $V_2O_5$ have low specific capacity. Finally, cathodes like sulfur and air are several years away from commercialization despite the widespread academic and industry interest.

Another possible cell configuration would be to build a cell with just copper foil as the anode. The lithium metal anode is plated out onto the copper foil during the first charge. This cell configuration, shown in figure 2m has no excess lithium leading to the highest energy densities. However, the lack of excess lithium leads to relatively low cycle life. Other advantages of this design are the need for only one webcoated electrode, no lithium metal, quick formation and the possibility to ship in the uncharged state.

Figure 3 shows the initial cycles of a cell with an anodeless design using a conventional Li-ion electrolyte. The cell delivers 2.2 Ah which is a 22% capacity improvement on the same cell design with graphite. The cell's thickness when charged is also 44% less than the graphite design. This leads to a large improvement in energy density. The pictures clearly show lithium deposits on the anode but the optical microscope images show the morphology is very grainy. Alternate electrolytes were explored to improve this morphology leading to better cycle life. A commercially available fluoroether was found to stabilize the cycling to >50 cycles when used at >80% solvent composition. This data is shown in figure 4. All data presented in the next section uses the anodeless cell design with a fluoroether based electrolyte.
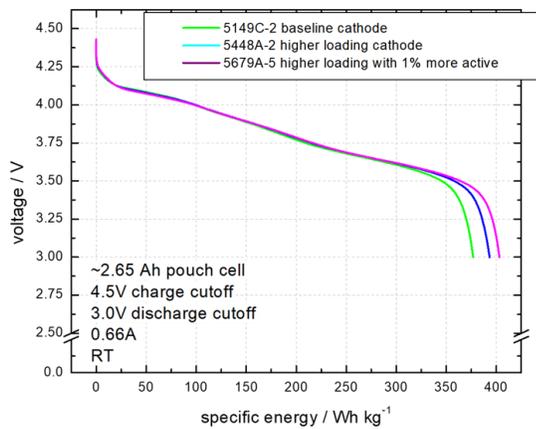


**Figure 3.** Pathfinder performance formation cycles of an anode less cell (left). Picture (top right) and optical microscope image (bottom right) of lithium plated on copper after 10 cycles.

**Figure 5.** Cathode development for EaglePicher's anodeless lithium metal cell.

## Anodeless Performance Data

The project this cell was developed on required a cell level specific energy >400 Wh kg$^{-1}$. More cathode active material must be added to this cell design to increase the capacity of the cell. This can be accomplished by increasing the cathode area (i.e. adding more cathode plates in a stacked pouch cell design), increasing the electrode loading and/or increasing the active percentage in a cathode formulation. All three methods were used for this project. Figure 5 shows the cell level specific energy was increased from 375 to 405 Wh kg$^{-1}$ by increasing the loading and active percentage.

The rate capability of the cell is shown in figure 6. The cell has an excellent rate capability for an energy cell delivering >300 Wh kg$^{-1}$ at a 6.6C continuous rate. The cell also was able to do >1 min 30A pulses. The rate capability is generally limited by the anode in a Li-ion cell. Since the discharge reaction for Li metal is a one electron oxidation it is very fast compared to a deintercalation process.
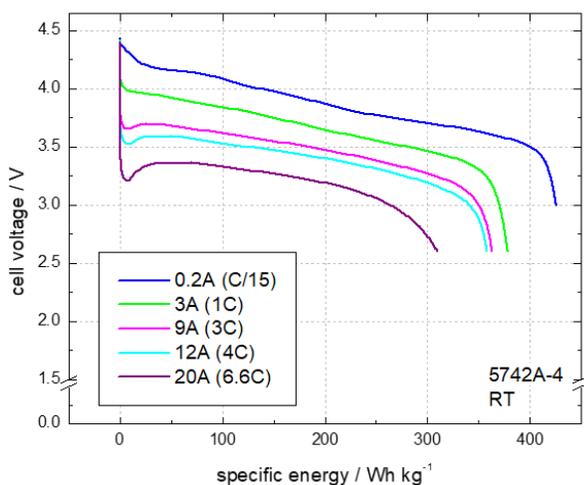


**Figure 6.** Rate capability of EaglePicher's anodeless lithium metal cell.
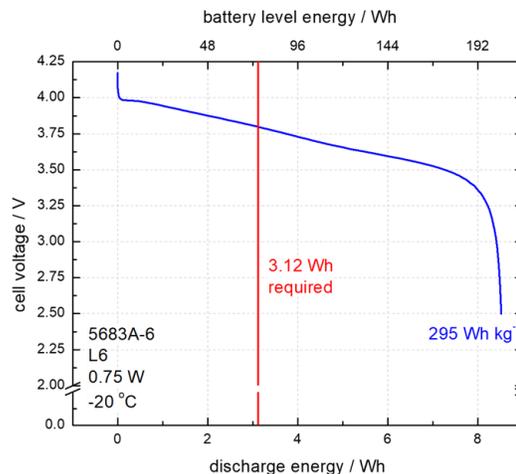


**Figure 7.** Cold discharge of anodeless lithium metal cell.

The cell was also able to discharge over a wide temperature range of −20 to +55 °C. Figure 7 shows the cell is able to deliver almost three times the required energy of the L6 Cold Discharge test in MIL-PRF-32383/4.

Figure 8 shows the cycle life of the cell. The cell is able to deliver 50-60 cycles consistently before its capacity retention <80%. New electrolytes and the addition of Li metal are being investigated to improve this cycle life. These will be discussed in the presentation.

## Conclusions

EaglePicher has developed a 3Ah rechargeable lithium metal cell with the anodeless cell design. The cell delivers >400 Wh kg−1 at a C/4 rate with excellent rate capability of 6.6C continuous. The cell is able to be discharged over a wide temperature range of −20 to +55 °C. Current efforts are underway to improve the cycle life to >125 cycles. Cell modeling shows specific energy >500 Wh kg$^{-1}$ is possible at larger cell form factors.
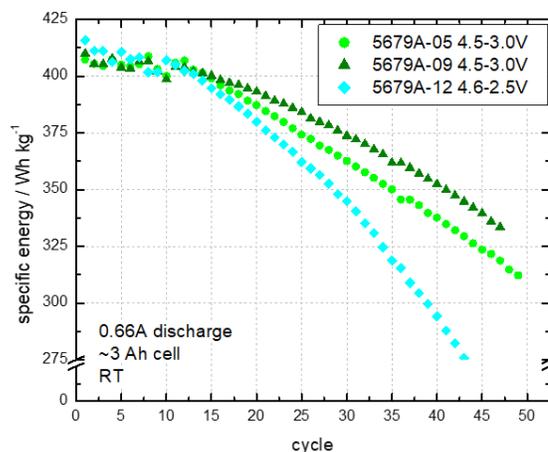


**Figure 8.** Cycle life of anodeless lithium metal cell.