

**Satellite Cell Development: Lithium-Ion Profile**

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**Abstract.** It is estimated since the latter part of the 1950's through the early 1970's, Eagle-Picher has manufactured over 1,000 nickel-cadmium (NiCd) spacecraft batteries and over 200 nickel-hydrogen (NiH<sub>2</sub>) spacecraft batteries. More recently (1994 to present), lithium-ion technology development has successfully produced and tested over 3,000 cells and several batteries. These high energy density, lithium secondary cells are very attractive for use on future spacecraft.

This paper summarizes the past and present activities of Eagle-Picher Technologies (EPT) in the quest to develop lithium-ion technology for spacecraft applications, as well as environmental issues and future development work. Specific technical challenges include the ability of lithium secondary cells to achieve the high cycle life and long calendar life required for use on high-reliability spacecraft, and the difficulties in scaling up the available small cell technology to larger sizes required for spacecraft power systems. With significant improvement over existing cell technology, the lithium-ion system will someday provide tremendous savings in lift-off cost as well as package flexibility.

**Introduction**

Eagle-Picher's baseline approach to lithium-ion electrochemistry, initially aimed at the aerospace and specialty markets, has developed at a rapid pace. The cell chemistry is the culmination of research including more than 100 carbon sources and types; greater than 15 cathode types from more than ten suppliers; and electrolyte investigations, both in-house and contracted. Cell design issues, performance, and cost are all factors in the choices made for testing.

The baseline design contains a LiCoO<sub>2</sub> cathode, graphitic type anode, ethylene carbonate based electrolyte, and polyethylene/polypropylene separator. The cells have been

configured in various ways dependent upon application and status of design (i.e., research-prototype-development).

**Technology Heritage**

Eagle-Picher has been a leader in batteries for spacecraft since the beginning of the industry's development. Having produced approximately 1,000 NiCd spacecraft batteries during the 1950's and 1960's and playing a significant role in the development of the NiH<sub>2</sub> system during the 1970's, Eagle-Picher set the stage for spacecraft battery production. Eagle-Picher has been the dominant producer of NiH<sub>2</sub> batteries throughout the 1980's and 1990's and has over 6,000 cells in 230 batteries powering over 130 satellites

currently in orbit. It is estimated Eagle-Picher batteries are used in greater than 85% of U.S. and international space programs utilizing NiH<sub>2</sub> technology.

This long history of activity in the U.S. and international space industry has resulted in the establishment of the world's most extensive spacecraft battery facility located in Joplin, Missouri. Joplin facilities include two separate satellite cell and battery manufacturing plants, capabilities for R&D to support existing and future development efforts, manufacturing engineering to support cell and battery production as well as in-house fabrication of many critical parts and sub-assemblies, complete test engineering, and an ISO 9000 quality system recognized by all U.S. and international prime contractors. Our services include, not only complete design and manufacturing support for our customer base, but extend support at contractor and launch facilities for integration, interface issues, and testing.

### **Lithium-Ion Heritage**

Eagle-Picher began evaluation of lithium-ion technology in 1994. IRAD development began in 1995 and contract development for the U.S. Government began in 1996. During

**Table I. Sample materials evaluated by EPT for Lithium-Ion Cells**

Component	Materials
Cathodes	LiCoO <sub>2</sub> , LiNi <sub>0.3</sub> Co <sub>0.7</sub> O <sub>2</sub> ,
	LiNiO <sub>2</sub> , LiNi <sub>0.2</sub> CoO <sub>2</sub> , LiMn <sub>2</sub> O <sub>4</sub> ,
	LiMn <sub>4</sub> O <sub>9</sub> , LiV <sub>2</sub> O <sub>5</sub> , LiV <sub>3</sub> O <sub>13</sub>
Anodes	KS-44, KS-15, KS-10, SFG-44,
	BG-39, SG-2933, RD8407, MM
	24-(XX), MM 6-28, MM 20-28,
	MM 6-10, MM 20-10, EC 300,
	EC 600, Vulcan XC72, MM 6-75
Electrolytes	EC/DEC, PC/DEC, EC/DMC,
	EC/DMC/DEC, EC/DMC/MA,
	LiPF <sub>6</sub> , MA/EC, LiBF <sub>4</sub>
Separators	Polyethylene, Polypropylene,
	PPP, PTFE, Ryton, Glass

this period, the following major lithium-ion cathode systems were evaluated: LiCoO<sub>2</sub>, LiNiO<sub>2</sub>, LiMn<sub>2</sub>O<sub>4</sub>, and the LiNi<sub>x</sub>Co<sub>y</sub>O<sub>2</sub> solid solution cathode system. A non-inclusive list of system elements investigated during this period is given in Table I.

During the first two years of this development period, EPT also investigated all of the appropriate methods of electrode processing. Applicability to the materials, physical dimensions, and dynamic requirements of current aerospace cells were evaluated. Cell development options were eventually narrowed to the LiCoO<sub>2</sub> cathode system. The electrodes were produced by either wet pasting (using TFE binders or web coating by doctor blade) or roll transfer (using PVDF binders). Both methods have been successful and data from both processes has shown strong cycle life capability within specific application ranges.

In late 1996, EPT separated the secondary lithium effort, developing a new operation, the Advanced Electrochemical Systems Operation. The main focus of the newly organized operation is development of liquid electrolyte lithium-ion systems.

During 1997, EPT IRAD efforts have included web coater process development using an enclosed coater capable of processing foil rolls six inches wide and up to 2,000 feet long. Coater development has resulted in routine production of double-sided electrode spools in thicknesses from twelve microns per side to 125 microns per side. Thickness tolerances on such double-sided electrode stock is five microns over the length of ten meters.

The combination of slurry development, coater, and in-house developed roll-mill processes now produces standard electrode

stock capable of supplying development cell needs from 4/3 A through 100 Ah sizes.

Current development of lithium-ion includes several important projects as follows:

- Demonstrate the feasibility of mid- and large-sized satellite cells.
- Investigate a new high capacity  $\text{Li}_x\text{Mn}_4\text{O}_9$  cathode material for special applications.
- Develop 4/3A cells for commercial applications.

Substantial EPT IRAD projects include increased coater slurry development, large satellite cell development (100Ah and larger), 28 volt, 15 Ah battery design improvements and full-scale evaluation, and large cell calorimetry.

The success of our lithium and nickel-based technologies in space applications are key elements in our development of lithium-ion and its adaptation to satellite design. Our existing lithium development capabilities have been used to extend conventional miniature lithium-ion performance to satellite cell demonstrations in 15 Ah sizes. This technology has also been demonstrated in 100 Ah cells. Feasibility of large lithium-ion satellite batteries is being expedited by our existing  $\text{NiH}_2$  satellite technology base. An improved low-weight spacecraft battery design is being used to produce the world's first aerospace quality 15 Ah, 28 volt lithium-ion satellite battery system.

This extensive technology heritage has resulted in the evolution of an optimized electrochemistry. Following is an indication of advanced performance achieved over the past months:

- High-stability, high-current-density electrodes for improved rate capability.
- Balanced electrolyte-to-active material ratio for extended cycle life.

- Optimized electrolyte composition for reduced impedance and increased temperature tolerance.
- Balanced cathode-to-anode capacity to insure proper Solid Electrolyte Interface (SEI) formation.

### Lithium-Ion System

Current cell development is divided into five areas:

- **Basic system development** in small test cells, both in bags and in stainless steel containers.
- **System/chemistry evaluation** in small spiral and prismatic stack cells.
- **15 Ah cell development** and demonstration in stainless and aluminum containers.
- **100 Ah cell development** and demonstration in stainless containers.
- **Mechanical optimization for mid- and large-size cells** to produce improved welding methods, aluminum containers, and header structures.

### **Basic System Development**

As the lithium-ion system was studied and adapted, it became clear that the system had opposing approaches available for cell design. The first was to design the cell strictly for energy density. The second was to design cells strictly for life. A third approach was a combination and optimization of the first and second. During the early stages of cell research, energy density goals of  $>125$  Wh/Kg were quite easily met with some cell designs providing more than 136 Wh/Kg. The problem lay in utilization of that material for extended periods of time at the rates necessary to support aerospace products. Fade rates increased dramatically between 60% and 100% utilization of materials. Refinement of

processing, charge regimes, particle distribution, conditioning procedures and material combinations all played heavily into the current design.

### System/Chemistry Evaluation

The EPT design chemistry has been placed in a variety of tests. Environmental characterizations have varied temperatures from 50° C to -40° C. Rate of charge and discharge have covered a broad spectrum as well: C/100 to 2C with pulses higher still. The follow-through on this effort will be the duplication of these tests with full scale-up 100 Ah cells.

The ability of cells to perform well over a broad range of power and temperature requirements presses a number of key variables to the extreme. Cornerstone variables of a broad-range system include the following aspects:

- **Electrolyte:** Must have acceptable conductivity at the lower temperatures as well as a known and defined degradation mechanism.
- **Cathode:** Must be stable in all operational settings ( $M^{2+}$  dissolution must be controlled or at least quantified). Rate capability and energy density are paramount.
- **Anode:** must maintain rate capability at the lowest of temperatures and have a slow self discharge mechanism at the highest of temperatures. Solid Electrolyte Interface (SEI) effects may be included here.
- **Processing:** Electrodes must be uniform in density and thickness (the more extreme

the temperature or power applications, the higher priority this area becomes).

- **Configuration:** Cells can be designed in various formations to accommodate the power and temperature extremes seen. Extremely low-temperature cells may need to be cylindrical to take advantage of the heat produced during a discharge. High-rate cells for space craft applications may need to be designed as prismatic stacks to take maximum advantage of the thermal dissipation along the axis of the current collectors.

### Cell Configuration

Figure 1 shows various cell designs that are currently in production. EPT has focused its attention on the large cells in a prismatic stack configuration. Spiral wound cells are also being built in 10 Ah sizes. Currently the pseudo-spiral wound, or folded configuration, has been evaluated in small cell sizes. At this time, EPT will not pursue further development of the pseudo-spiral wound configuration due to electrical and mechanical problems with this design. Development efforts will undoubtedly pursue larger cell sizes in the stacked configuration.

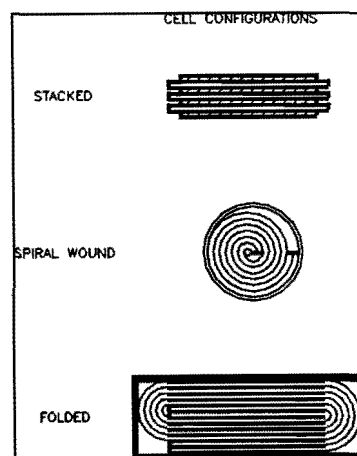


Figure 1. Cell Configurations

The selection of the cell configuration is centered around cycle life of the system, and the thermal management of that system. Thermal management has developed into a key issue with the lithium-ion designs for satellite application. The choices for configuration can also be influenced to a lesser degree by packaging efficiencies, package flexibility, and ease of manufacture, all of which lead to cost saving efforts for the satellite community. The basic chemistry of lithium-ion creates five main problems for the satellite cell designer: thermal dissipation, mechanical stress, packaging, cell scalability, and uniformity. The main challenge is the expansion and contraction of the materials upon charge and discharge.

### *Thermal Dissipation*

Systems which do not dissipate or distribute heat well are doomed to shortened lives by increased electrolyte degradation at what are termed "hot spots" in the system. Heat imbalance in the lithium-ion system tends to force better performance at the elevated temperature portion of the cell, but results in increased fade and increased incidence of cell shorting due to a higher probability of plating lithium materials.

The lithium-ion system in standard form, that is Cu current collector for the anode and Al collector for the cathode, tends to transfer heat in a path parallel to the electrodes utilizing the high heat transfer capabilities of the Al, Cu and carbon materials. Heat dissipation in the direction perpendicular to the plates is limited by the insulating qualities of the  $\text{LiCoO}_2$  material. With heat dissipation parallel to the electrodes, the obvious design for maximum thermal control (cycle life) in aerospace situations will be the prismatic stack. Although the spiral and pseudo-spiral will perform adequately in convection

atmospheres, their usefulness in orbiting space craft with high rate requirements may be limited. Reported data shows large temperature differences between the heat-dissipating ends of a spiral-wound can and the outside, lateral portion of the can. Some of these numbers have been reported as high as  $\Delta 50^\circ \text{C}$ .

### *Mechanical Stress*

During the charge process, cathode material may expand from three to six percent dependent upon type, density, thickness, and surface area. The anode also expands during the charge process from 14% to more than 30%, dependent upon the type, density, thickness, and particle surface area. Controlling this expansion in a spiral-wound system is relatively easy in that the system will expand and move to the center of the cell arrangement. The prismatic stack on the other hand has no self preserving mechanism to maintain the constant pressure required by the system. In this case, external forces need to be applied. This was one of the major battles during the birth of the lithium-ion system at Eagle-Picher. Many techniques were tried with very limited success until the realization that the electrode materials can be utilized to apply the pressure by plate and stack design. The pseudo-spiral-wound type of cell technology contains the best of both worlds, as well as the worst of both worlds. The pseudo-spiral-wound technology allows expansion in the stacked area as well shear force to the plate in the corner of the cell. Although, through design of the casing and cell ratio H/W/L, the expansion can be lowered. Long-term cycle life may see current differences across the electrode as well as increased micro-mechanical damage in this design.

### Packaging

Certain cell configurations lend themselves to package flexibility. The spiral-wound cell, while higher in energy density at the cell level, may lose this advantage at the battery level. Current projections show spiral-wound cells losing more than 24% to packaging while the same size cell in the prismatic stack design loses only 10-12% to packaging. Utilizing the prismatic stack system, cell design can be built into the end product allowing for proper thermal transfer in a multitude of shapes and sizes.

### Cell Scalability

Utilizing thermal modeling and other mechanical design tools, a direct scale-up has been accomplished from the existing 15 Ah size to the 100 Ah size. The diagrams below (Figures 2, 3, and 4) show the dimensions of the current 15 Ah and 100 Ah cells and the electrode design.

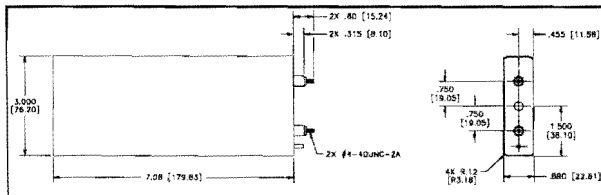


Figure 2. Drawing of 15 Ah Cell

Attention should be given to the electrode stack design. The same electrode design is currently being used in both cell types. Direct scale-up is possible with the prismatic stack system. Problems associated with the spiral-wound system for the most part stem from heat build up in the center of the cell causing a difference in material utilization and life. This problem becomes even more apparent with larger cell sizes.

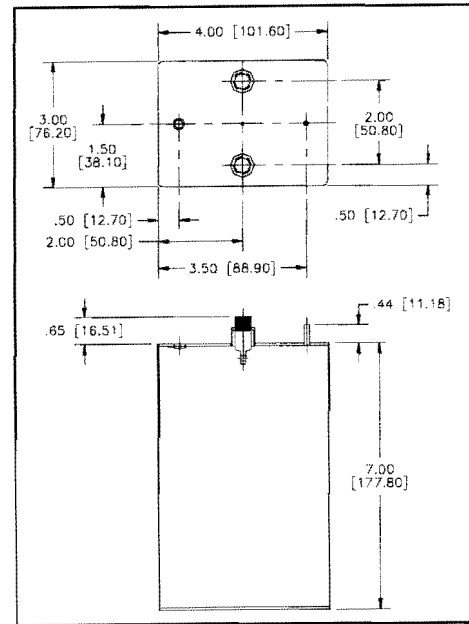


Figure 3. Drawing of 100 Ah Cell

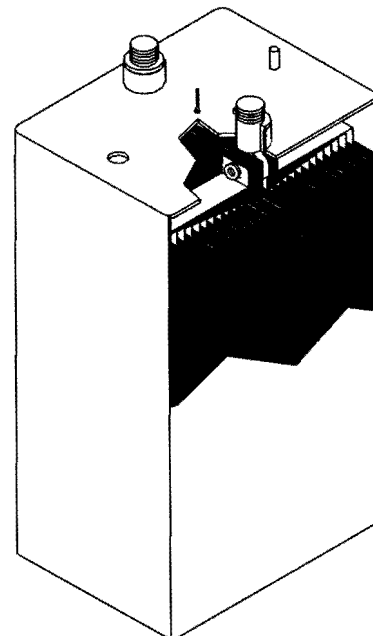


Figure 4. Cross-sectional view of 100 Ah cell

### Uniformity

With any system, uniformity between cells dictates how the battery will operate under given conditions. A battery system will only be as good as it's weakest cell. The table

below (Table II) defines differences in a typical cell lot after conditioning. At this point in the process there has been no electrode or cell matching. This uniformity is well maintained. Cell divergence of only 4% in 100 cycles would be considered acceptable or even exceptional in most applications. However the satellite industry requires greater than 40,000 cycles for a standard LEO application. The ability of this system to perform well without electronics to control individual cells is still under investigation. At this point Eagle-Picher is conducting full-scale battery testing and data to date looks promising for series connected batteries with limited cell charge control. The implementation of series cells for lithium-ion battery applications in space is awaiting more defined requirements. Eagle-Picher has discussed with many customers that perhaps for the first generation of lithium-ion batteries in space, individual cell charge control will be necessary. Future generations may not require this electronic structure, but further proof is needed to verify these claims.

**Table II Cell capacity performance after 125 cycles.**

Cell Capacity Performance in Modified GEO Regime		
Cell Number	Discharge Capacity (Ah)	Cycle 125
2739702	12.97578	<b>Mean Capacity</b> 12.39061 <b>Standard Deviation (mAh)</b> 11.00107 <b>COV</b> 5.27% <b>% Variation</b> 4.72%
2749701	12.96557	
2769701	12.77098	
3079701	12.92741	
3089702	11.3106	
3099701	11.56827	
3099702	12.21768	

### 15 Ah Cell Development

The EPT baseline 15 Ah cell design technology is outlined in Table III. Currently, our production team is producing two to five 15 Ah cells per day. The continuous production will aid in optimization of manufacturing methods, development for future production and will produce cells in

quantity for expanding characterization, additional battery production demonstrations and hardware samples. Ongoing cell evaluation includes testing at various rates, temperatures, and DODs; characterization; capacity fade on stand to include effects versus state of charge, as well as temperature; and LEO and GEO life testing.

**Table III. Summary of cell design**

DESIGN SUMMARY SHEET	
15 Ah CELL	
Capacity (Ah)	> 16 Ah
Electrolyte	cyclic organic
Separator	porous polypropelene
Rated Capacity	15 Ah
Limited by	Anode
Wh/Kg Cell Level Rated	> 80

### Cell Capabilities

EPT lithium-ion current capabilities (Table IV) show the diversity of testing and cell designs we are now encompassing in EPT's baseline.

**Table IV. Cell Capabilities.**

Eagle-Picher Lithium-Ion Current Capabilities (as of 7/15/98)	
Continuous Discharge Capability	C (Pulse Rates Higher)
Operational Temperatures	-20°C ---> +50°C
	>830 @ 100% DOD @ C/2 Discharge
	>1040 @ 50% DOD @ C/2 Discharge
Cycle Life	>1120 @ 42% DOD @ C/5 Discharge & -4° C
	>2780 @ 25% DOD @ C/2 Discharge
	>3220 @ 14% DOD @ C/5 Discharge & -4° C
	>12600 @ 10% DOD @ C Discharge
Wh/Kg	>80 @ C rate (have achieved >135 Wh/Kg)
Wh/l	>235 @ C rate (have achieved >290 Wh/l)
Shelf Life	Test in Progress
Cell Charge Control	CC, CV EP currently designing a battery baseline.
Cell Sizes	20 mAh - 100 Ah (400 Ah sizes on drawing board)
	Prismatic Stack
Designs	Spiral Wound
	Pseudo Spiral Wound

Including the standard battery of tests, these capabilities allow for electrical testing of both cells and batteries for qualification and acceptance purposes. Beyond electrical testing, we have the capability to perform vibration, shock and other environmental simulation testing to a wide range of requirements.

### Coefficient of Variance

The Coefficient of Variance graph (Figure 5), shows the difference in capacity spread of cells after conditioning during the '96-'97 time period. For fiscal year 1996, a different web coating

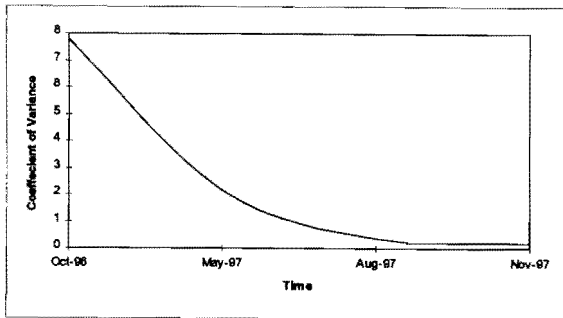


Figure 5. Coefficient of Variance

process was used to produce most of the cell designs. As we moved into the year of 1997 and began to automate our processing, our COV decreased rapidly. Cell capacity spread at 100% cycling, is extremely tight, having a COV of only 0.2%. The close capacity and impedance tolerance implied by this data, despite the preliminary design and manufacturing methodologies, validates our belief that this cell system can be produced in typical satellite battery sized cells and with reproducibility meeting and possibly surpassing the NiH<sub>2</sub> system.

### Recharge Ratio

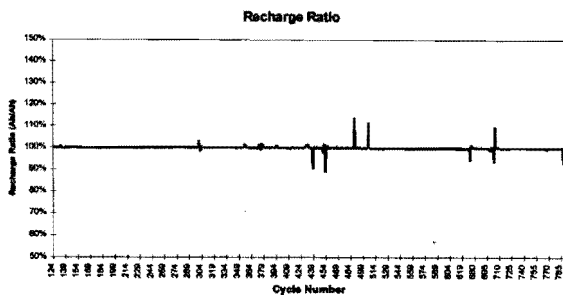


Figure 6. Recharge ratio

Figure 6 shows the recharge ratio of a standard 15 Ah cell, cycling at a C/5 charge, C/2 discharge. This is a modified GEO type orbit at 100% DOD.

### Cycle Life

Projected cycle life for cells at varying DODs is shown in Figure 7.

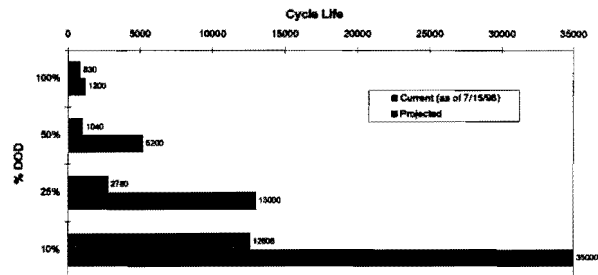


Figure 7. Cycle life characteristics - present and projected

Today the 10% cells are out to 12,600 cycles with projected cycle life of >35,000. Twenty-five percent DOD cells are out to 2780 with a projected cycle life of >13,000. Fifty percent DOD cells are out to 1040 with a projected cycle life of >5,200. One hundred percent DOD cells are out to 830 with a projected cycle life of 2000.

The improved cell technology for 1997 has shown a marked improvement in part due to the implementation of better quality systems and handling procedures. Expected performance and cycle life improvements may be as high as one and one-half times current projections.

### 100 Ah Cell Development

Eagle-Picher's designs are not limited to the smaller 15 Ah cells. As stated earlier, the development of a true prismatic stack system has led to a direct scaled up version of the 15 Ah cell to a 100 Ah cell. The design of the 15



Ah and the 100 Ah cells are very similar, the same electrodes are used in both cases.

The 100 Ah cell contains a larger surface area for heat dissipation and larger terminal designs, which are the key areas of difference between the two designs. This scalability, when combined with the very favorable heat dissipation qualities of the prismatic stack system, are the deciding factors in Eagle-Picher's configuration choice.

EPT produced the first 100 Ah cells during 1997. These cells are currently being tested. A small number of cells will be produced over the next six months to allow preliminary characterization of the cells and provide feasibility data for further development.

### Mechanical Optimization

As with the nickel hydrogen system, the structural and thermal hardware that comprises the battery is a sophisticated system of aluminum alloys, RTV's, thermal transfer enhancement materials, and high strength fastening systems employed in an electrically neutral architecture.

### Battery Development

#### 28V, 15 Ah Battery

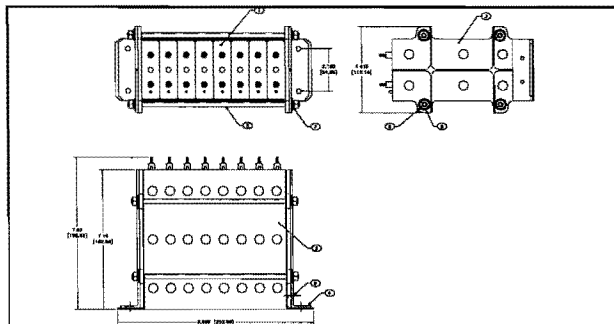


Figure 8. Drawing of 28V, 15 Ah battery

The first 28V, 15 Ah batteries intended for satellite performance demonstration were produced in 1997. Eight cells of the current 15 Ah design, were placed in a battery configuration (Figure 8) with a lightweight aluminum casing. Battery configuration, performance, and development are currently under evaluation.

### Goals

For the ultimate application of lithium secondary batteries to high reliability spacecraft, the following operational goals have been proposed:

#### Cycle Life:

- 2500 cycles in GEO orbit @ 70% DOD
- 35,000 cycles in LEO orbit @ 40% DOD

#### Calendar Life:

- 10 yr. on orbit plus. 3 yr. storage

#### Specific Energy:

- 125 W hr/kg (cell level-total)
- 110 W hr/kg (battery level-total)

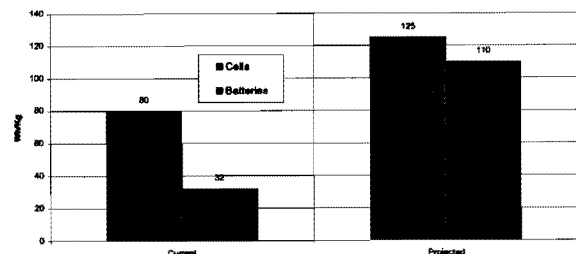
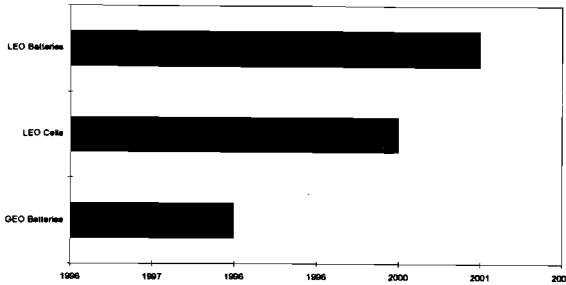


Figure 9. Current and Projected Specific Energy (Wh/Kg)

Figure 9 shows the current progress vs. projected goals of specific energy at the cell and battery levels.



**Figure 10. Development Timetable**

Figure 10 illustrates the timetable set for cell and battery development.

### Future Development

Future applications of the lithium-ion system will cover a broad range of temperature, power, and cycle life requirements. Systems projected to date have covered temperature ranges from  $-80^{\circ}\text{C}$  to  $+80^{\circ}\text{C}$ . Power requirements have varied from  $300\mu\text{W}$  to 120 KW. Cycle life has varied from 50 cycles to 40,000 cycles.

Continued system development will focus on cycle life enhancements and energy density improvements, both intended to maintain our pace of development over the next few years. Cycle life issues include corrosion studies, electrolyte purity issues, and voltage limit effects. Energy and power issues include mechanical improvements, continued electro-processing issues and raw materials issues. Table V highlights future development plans.

These projects will be performed under the planned EPT IRAD as part of anticipated government and industry contracts. We also anticipate a continued expansion of our facilities and equipment base due to parallel development of both GEO and LEO designs in larger cell sizes as well as expansion of the effort to include alternative processing methods such as a plastic-based systems.

**Table V. Development plans for the near future**

Expanded definition of 15 Ah cell performance
Production and test of 100 Ah cells
Performance and thermal evaluation of a 28 volt - 15 Ah battery
Continued system development including electrolyte and material improvements
Continued mechanical development including aluminum containers for increased specific energy
Specific IRAD projects including development of new SnO materials through a teaming partner.

### Summary

Test data on the current technology of web coated 15 Ah cells is incomplete. The cells are still cycling at various profiles and appear to be only partially into their cycle lifetime. Cycling has been emphasized at 100% high rate discharge to stress the system and reduce test time. We have recently begun low DOD testing and are now developing methods for accelerated life testing.

We can, however, make a number of inferences from existing data. The EPT chemistry is capable of C rates and has shown promise in cycle life. Operational temperature testing has shown abilities at various temperatures between  $-20^{\circ}\text{C}$  and  $+50^{\circ}\text{C}$ . Variations in cell capacity among lots has shown marked improvement with the addition of automated processing equipment. Energy densities of 80 Wh/kg to 120 Wh/kg will have significant impact on the aerospace systems of the future.

Although flight heritage is extremely important, lithium-ion secondary rechargeable batteries have yet to fly a mission in space. It is the intent of Eagle-Picher through in-house design and qualification efforts, to take

existing nickel hydrogen technology and combine current lithium-ion cell development efforts to fabricate a feasible flight battery.

Many technical challenges exist before lithium-ion technology will have the reliability and knowledge base necessary to provide primary satellite power. We believe however, that current levels of performance, our rapid development rate, and existing base of lithium and NiH<sub>2</sub> spacecraft technologies, will make EPT successful in development of the system for space applications. These existing capabilities should provide for rapid development, rapid adaptation of spacecraft quality and reliability, and rapid manufacturing startup.

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