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A Study on Advanced Lithium-Based Battery Cell Chemistries to Enhance Lunar Exploration Missions

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Abstract

NASA's Exploration Technology Development Program (ETDP) Energy Storage Project conducted an advanced lithium-based battery chemistry feasibility study to determine the best advanced chemistry to develop for the Altair Lunar Lander and the Extravehicular Activities (EVA) advanced lunar surface spacesuit. These customers require safe, reliable energy storage systems with extremely high specific energy as compared to today's state-of-the-art (SOA) batteries. The specific energy goals for the development project are 220 watt-hours per kilogram (Wh/kg) delivered at the battery-level at 0 degrees Celsius (°C) at a C/10 discharge rate. Continuous discharge rates between C/5 and C/2, operation between 0 and 30 °C, and 200 cycles are targeted.

The team, consisting of members from the NASA Glenn Research Center, the NASA Johnson Space Center, and the NASA Jet Propulsion Laboratory, surveyed the literature, compiled information on recent materials developments, and consulted with other battery experts in the community to identify advanced battery materials that might be capable of achieving the desired results with further development. A variety of electrode materials were considered, including layered metal oxides, spinel oxides, and olivine-type cathode materials, and lithium metal, lithium alloy, and silicon-based composite anode materials. Lithium-sulfur systems were also considered. Hypothetical cell constructs that combined compatible anode and cathode materials with suitable electrolytes, separators, current collectors, headers, and cell enclosures were modeled.

The relative safety of systems containing the materials under consideration was assessed. Risks were also factored into the decision making process. The risks include uncertainties due to ease of scaling-up of large batches of raw materials, adaptability of the materials to processing using established or reasonable cost manufacturing techniques, manufacturability of the materials in dimensions required for integration into battery cells of practical capacities, Technology Readiness Levels (TRL), and the likelihood of achieving the desired performance by the customer need dates. The advanced cell chemistry options were evaluated with respect to multiple quantitative and qualitative attributes while considering their projected performance at the end of the available development timeframe. Following a rigorous ranking process, a chemistry that combines a lithiated nickel manganese cobalt oxide $Li(LiNMC)O_2$ cathode with a silicon-based composite anode was selected as the technology that can potentially offer the best combination of safety, specific energy, energy density, and likelihood of success.

Tasks over the next 3 years will focus on development of electrode materials, compatible electrolytes, and separator materials, and integration of promising components to assess their combined performance in working cells. Cells of the chosen chemistry will be developed to TRL 6 by 2014 and will then be transferred to the customers for infusion into their mission paths.

Introduction

NASA's Constellation program is designing and building a new fleet of vehicles which will enable a permanent human presence on the Moon. Safe, lightweight, and compact energy storage technologies are required to enable or enhance these vehicles. In recent years, state-of-the-art (SOA) lithium-ion (Li-ion) cell technology has revolutionized the energy storage capabilities of space-rated batteries by enabling mission applications that require low temperature batteries, such as the 2003 Mars Exploration Rovers (MER), the Mars Phoenix Lander Scout Mission and the Mars Science Laboratory. Li-ion batteries have also significantly enhanced the capabilities of Earth-orbiting satellites and astronaut portable power applications, such as Personal Digital Assistants, by offering 2 to 3 times lower mass and volume energy storage systems as compared to traditional nickel-based spacecraft batteries.

The next generation of space vehicles will require even lighter-weight energy storage systems. Additionally, unlike prior aerospace missions that have utilized Li-ion batteries, Constellation missions will require the combination of an energy storage system that will provide primary power for the vehicle and the increased level of safety befitting a human-rated mission. Existing SOA battery technology does not possess the capability to address all of the mission requirements. NASA's ETDP Energy Storage Project is developing advanced Li-ion batteries for Altair Lunar Lander, EVA, and Mobility Systems for Lunar Surface Systems (LSS) with the goal to provide the best combination of safety and low mass in a compact system to address their mission requirements.

Customer Requirements

The ETDP Energy Storage Project is developing two Li-ion cell chemistries to address customer requirements, the high energy (HE) cell and the ultra high energy (UHE) cell. Among the goals of both cell development activities are increased safety, specific energy, and energy density over SOA aerospace Li-ion cell technology.

Mobility Systems for LSS include pressurized and unpressurized rovers and habitats. These vehicles have a wide range of requirements that are still being defined. Based on the preliminary requirements for Mobility Systems, a HE cell development effort was formulated to address the LSS customer requirements. The objective is to enable a battery system that can operationally deliver 150 Wh/kg at the beginning of life and can provide 2000 cycles to 80 percent of its original capacity. To accomplish these goals, cathode, electrolyte, and safety components are being developed and advanced separators are being assessed for HE cells. The HE cell goals represent increased safety over SOA Li-ion technology. Advanced cathode development for these cells is expected to enable over a 50 percent gain in cell-level specific energy over SOA.

The UHE cell development is targeted for the ascent stage of Altair, and to power the Portable Life Support System (PLSS) for the EVA Lunar spacesuit. For these missions, mass is highly critical, but only a limited number of cycles are required. EVA desired upwards of 300 Wh/kg of useable energy for the mission (i.e., delivered on a battery level), far beyond the capabilities of existing technology. Cathode-level specific capacity enhancements alone were incapable of increasing the cell-level specific energy enough to meet the customers' requirements, therefore anode materials with higher theoretical specific capacity than conventional graphitic anode materials had to be employed to enable the UHE cell. The low cycle life requirement allowed for the pursuit of different classes of anode materials that have demonstrated promising specific capacity performance results, but whose limited cycle life potential has inhibited much of the interest in these materials for commercial development.

Attributes

Determination of the best potential chemistry to develop to enable an UHE system to address EVA and Altair's requirements was the focus of this feasibility study. Ten attributes were chosen to rank each of the chemistries. These attributes represent the figures of merit that were determined to be the most important to the final goal of choosing an advanced chemistry that has the best combination of safety, specific energy, energy density, and likelihood of success. The ten attributes are: cost to TRL 6, cycle life, energy density, manufacturability, rate capability up to C/5, rate capability up to C/2, safety, schedule, specific energy, and storage and calendar life. Several attributes were deemed to contribute to likelihood of success, including manufacturability, cycle life, cost to TRL 6, schedule, and rate capability. These attributes were each considered individually. The attributes and their definitions are given in Table 1.

TABLE 1.—ATTRIBUTES FOR RANKING CHEMISTRIES AND THEIR DEFINITIONS

Attribute	Definitions
Cost to TRL 6	The cost to develop the technology to TRL 6, including costs attributed to costly manufacturing processes or processes that cannot be automated
Cycle life	Projected cycle life of the technology
Energy density	Projected energy density of the technology (calculated under a standard set of conditions)
Manufacturability	The projected level of ease or difficulty associated with working with materials, scaling up batches of materials, and manufacturing cells of practical capacity made from these components, and the projected adaptability of materials to large scale processing
Rate capability up to C/5	Likelihood that the technology can meet a C/5 continuous discharge rate
Rate capability up to C/2	Likelihood that the technology can meet a C/2 continuous discharge rate
Safety	The likelihood that a cell made from these components can be made to be safe. Included safety under normal operation and abuse conditions
Schedule	Likelihood that TRL 6 cells can be delivered by March 2104
Specific energy	Projected specific energy of the technology (calculated under a standard set of conditions)
Storage and calendar life	Projected storage + calendar life, where calendar life includes the operating time plus periods at open circuit between active charging and discharging

Chemistry Options

The chemistries that were considered were selected as a result of extensive literature surveys, compilation of information on recent materials developments, and consultations with other battery experts in the community to identify advanced battery materials that might be capable of achieving the desired results with further development. A variety of electrode materials were considered, including layered metal oxides, spinel oxides and olivine-type cathode materials, and Li metal, Li alloy and silicon (Si)-based composite anode materials. Li-sulfur (Li/S) systems were also considered.

Of the numerous options that were considered, options for detailed consideration were narrowed down to 32 choices. Hypothetical cell constructs that combined compatible anode and cathode materials with suitable electrolytes, separators, current collectors, headers, and cell enclosures were modeled using a battery model developed under the ETDP Energy Storage Project. The outputs of the model were projections on cell and battery-level specific energy and energy density for the different options under consideration. These results represent projected performance after 3 years of focused component development.

A 35 Ah cell was assumed to approximate specific energy and energy density for a cell of practical capacity for the customers and to form a common basis of comparison of chemistries. Two different cell geometries were considered, prismatic (rectangular) and cylindrical cells. The results for the 32 chemistries are shown in Figure 1 and Figure 2. The 300 mAh/g ETDP cases are chemistries that use the projected performance of the $Li(LiNMC)O_2$ cathode material currently being developed under the ETDP project, where LiNMC is $Li_xNi_yMn_zCo_{1-x-y-z}$. 300 mAh/g is the projected room temperature specific capacity of the materials.

As seen from the differences in the specific energy and energy density results between the prismatic and cylindrical cell designs, cell designs can have a significant effect on mass and volume. Four cell chemistries were chosen to illustrate these effects. Specific energies for a 35 Ah prismatic cell in a stainless steel can with a 20-mil wall thickness, a 35 Ah cylindrical cell in an aluminum can with a 60-mil wall thickness, a 35 Ah prismatic cell in a plastic case with a 20-mil wall thickness and an 18650-size cell were calculated. An 18650 cell is a cylindrical cell form factor with an 18 mm diameter and a 65 mm height and is commonly used for commercial cells.

As seen in Figure 3, the specific energy of a cell chemistry can increase by as much as 100 Wh/kg when only the cell packaging is considered. Cell packaging must be combined with appropriate battery packaging to truly see the gains achieved by lightweight cell construction materials. In the case of plastic prismatic cells for instance, many of the cell-level specific energy gains may be lost in packaging a flight-quality battery composed of cells using these packaging materials. Appropriate cell designs, including packaging that can enable the lightest weight flight battery system, will be determined as the development activity progresses.

Since it was not practical to perform a detailed weighting analysis on each of these chemistries, several options were eliminated on the basis that their projected specific energy did not come close to achieving the customers' goals. A threshold was drawn at 180 Wh/kg. Chemistries that clearly did not come close to the threshold when packaged in either prismatic or cylindrical metal cases were eliminated. High voltage cathode/anode combinations, lithium titanate chemistries, and chemistries that used the SOA meso-carbon microbead (MCMB) anode did not meet the minimum threshold specific energy. Plastic prismatic case packaging was eliminated since the gains here were not related to electrochemical advancements and each of the chemistries that were modeled with plastic were considered elsewhere with other packaging. Of the remaining classes of materials, seven specific chemistries were chosen for the detailed weighting analysis. These options and their descriptions are listed in Table 2. Specific energy and energy density predictions for these seven options are shown in Figure 4 and Figure 5 and their percent gain in specific energy over SOA Li-ion cells is shown in Figure 6.

TABLE 2.—FINAL CHEMISTRY OPTIONS

Anode	Cathode	Description
Si-Composite	$Li(LiNMC)O_2$ (ETDP)	Si-based composite anode with 1000 mAh/g specific capacity and 14% irreversible capacity. Lithiated nickel manganese cobalt oxide layered cathode with 300 mAh/g specific capacity and 4 mil electrode thickness. Cathode currently under development for the ETDP Energy Storage project. LiNMC is $Li_xNi_yMn_zCo_{1-x-y-z}$.
Si-Composite	$Li(Ni_{0.33}Mn_{0.33}Co_{0.33})O_2$	Si-based composite anode with 1000 mAh/g specific capacity and 14% irreversible capacity. Nickel manganese cobalt oxide cathode (commercial "one third, one third, one third" formulation).
Li-metal	$Li(Ni_{0.33}Mn_{0.33}Co_{0.33})O_2$	Li-metal anode. Nickel manganese cobalt oxide cathode (commercial "one third, one third, one third" formulation).
Li-metal	$Li(LiNMC)O_2$ (ETDP)	Li-metal anode. Lithiated nickel manganese cobalt oxide layered cathode with 300 mAh/g specific capacity and 4 mil thick electrode. Cathode currently under development for the ETDP Energy Storage project. LiNMC is $Li_xNi_yMn_zCo_{1-x-y-z}$.
Li-metal	$LiNiMn_2O_4$	Li-metal anode. $LiNiMn_2O_4$ cathode, spinel structure.
Li-metal	$LiCoPO_4$	Li-metal anode. $LiCoPO_4$, olivine structure.
Li-metal	$(Li_2)S$	Li-metal anode. Sulfur cathode with 1100 mAh/g specific capacity and 25% diluent.

Ranking Process

The Analytic Hierarchy Process (AHP) was chosen as the decision making tool for the feasibility study (Ref. 1). This process allowed the team to generate weightings in a stepwise fashion for each attribute with respect to every other attribute and for each chemistry with respect to each attribute. The intermediate results were then combined to generate the overall weightings for the attributes and for the chemistries. The chemistry which resulted in the highest weight is the preferred chemistry. The results of the ranking process are discussed in the following sections.

Weighting of Attributes With Respect to Choosing an Advanced Chemistry

The process of weighing each attribute with respect to its importance in choosing an advanced chemistry resulted in the weightings shown in Table 3. Safety rose as the top priority with a final weight of 17.9. This weighting is consistent with the customers' slight preference for safety over performance. Rate capability to C/5 and specific energy were weighted closely together as the second and third priorities. A post study sensitivity analysis showed that the final study results were unaffected by switching around the weights of the top three attributes. These results reflect the customers' requirement for extremely light batteries that can meet the mission requirements. Based on the customers' present load profiles, low to moderate discharge rates are required.

TABLE 3.—ATTRIBUTE WEIGHTINGS WITH RESPECT TO CHOOSING AN ADVANCED CHEMISTRY

Attribute	Final weight
Safety	17.9
Rate capability up to C/5	15.6
Specific energy	15.0
Storage and calendar life	12.2
Energy density	10.2
Manufacturability	8.3
Schedule	8.0
Cost to TRL 6	6.5
Cycle life	3.8
Rate capability up to C/2	2.5

Rate capability up to C/2 was ranked as the least important attribute since there is no existing specific requirement for a C/2 discharge rate. There is, however, an expectation that load profiles will grow as requirements change until the final design is set. An additional driver for the consideration of an increased discharge rate capability is the highly critical nature of the mass requirement for the EVA customer.

Although the EVA customer overwhelmingly prefers a battery that can perform for the entire duration of an 8-hr sortie, in the event that the battery mass is too prohibitive for an astronaut to practically carry, the customer may be willing to sacrifice battery discharge time in order to obtain a lighter battery. A battery that operates for 4 hr will save approximately half the mass of an 8-hr battery and can be swapped with a spare midway through the sortie to still allow for an 8-hr sortie. The reduction in energy in the battery will result in an increased discharge rate requirement since the same amount of power will be needed to meet the mission requirements as in the 8-hr case, therefore a maximum discharge rate of C/5 will no longer be adequate to meet the load profile.

Due to these considerations, a conservative estimate of C/2 was deemed as the maximum possible discharge rate, but its importance in choosing an advanced chemistry was not judged to be very significant. The next sections will discuss the weightings of each of the seven chemistries with respect to each of the ten attributes.

Weighting of Chemistry Options with Respect to Attributes

Safety

The integration of all of the components that make up a cell contribute to its overall safety. For this study, the individual contributions of the anode and cathode and their potential impact on the safety of each chemistry were considered. While safer electrolytes and separators can increase the overall safety of a cell, for simplicity, the impacts due to these components were assumed to be similar for all chemistries and did not factor into the safety rankings. The resultant weightings solely reflect the ranking of the perceived safety of the anode/cathode combinations for each chemistry. Cell-level safety was considered for normal operation and under abuse conditions.

For the seven chemistries, there were two choices of anodes: Li-metal and Si-based composite. There are serious safety concerns associated with a rechargeable Li-metal chemistry (Refs. 2 and 3). As a cell that contains a Li-metal anode cycles, it becomes prone to Li dendrite growth with each additional cycle as Li is deposited unevenly on the electrode surface. The dendrites can puncture the separator and cause internal shorting (Ref. 4). Coating the surface of Li-metal anodes could inhibit dendrite growth (Ref. 5), however, the development of appropriate coatings in the available timeframe will pose a significant technical challenge. Without the appropriate coatings, Li-metal rechargeable chemistries cannot meet an acceptable level of safety. For these reasons, all Li-metal chemistries received weightings that were significantly lower in safety than Si-based composite anode chemistries.

The safety of the cathode materials was determined based on their inherent thermal stability and voltage capability. The $Li(Ni_{0.33}Mn0_{.33}Co_{0.33})O_2$ and $Li(LiNMC)O_2$ ETDP cathode materials are characterized by higher thermal stability than conventional Li-ion cathodes, which is attributed to their Mn content (Refs. 6 to 8). The $Li(LiNMC)O_2$ ETDP cathode materials are capable of operating at high voltages (above 4.5 V) as compared to conventional Li-ion cathodes (~4.2 V). When packaged in the same physically-sized cells, a cell containing the $Li(LiNMC)O_2$ ETDP cathode would have a higher energy content than a cell containing a conventional cathode, and would thus pose a greater safety concern under abuse conditions than a lower energy cell. When paired with the safer anode material, the higher voltage operation of the $Li(LiNMC)O_2$ ETDP cathode was deemed to outweigh its thermal stability properties as a higher safety risk, which resulted in the Si-based composite/ $Li(Ni_{0.33}Mn0_{.33}Co_{0.33})O_2$ cathode chemistry receiving the highest weight of 38.9 for Safety. When paired with a more volatile Li-metal anode, however, the cathode became less important in the determination of safety, resulting in similar weights for all of the Li-metal chemistries. The results of the Safety rankings are shown in Table 4. Figure 7 effectively illustrates the impact the dominance of the Si-based composite anodes on the safety of the chemistry.

TABLE 4.—WEIGHTING OF CHEMISTRY OPTIONS WITH RESPECT TO SAFETY

Anode	Cathode	Weight
Si-Composite	$Li(LiNMC)O_2$ (ETDP)	28.8
Si-Composite	$Li(Ni_{0.33}Mn_{0.33}Co_{0.33})O_2$	38.9
Li-metal	$Li(Ni_{0.33}Mn_{0.33}Co_{0.33})O_2$	6.3
Li-metal	$Li(LiNMC)O_2$ (ETDP)	8.4
Li-metal	$LiNiMn_2O_4$	5.7
Li-metal	$LiCoPO_4$	5.7
Li-metal	$(Li_2)S$	6.1

Rate Capability up to C/5

When considering rate capability to C/5, it was again necessary to compare the performance of individual electrodes that impact rate in addition to the anode/cathode couples. The $Li(LiNMC)O_2$ ETDP cathode currently has known rate limitations as compared to $Li(Ni_{0.33}Mn_{0.33}Co_{0.33})O_2$ cathode, so $Li(Ni_{0.33}Mn_{0.33}Co_{0.33})O_2$ cathode ranked higher than the $Li(LiNMC)O_2$ cathode when paired with either anode.

There is not much information available in the literature regarding the rate capability of Si-based composite anode materials, however, they are assumed to exhibit poorer rate capability than Li-metal anodes. Chemistries containing Si-based composite anodes are generally weighted lower than those containing Li-metal anodes.

The sole exception is the $LiCoPO_4$ cathode, which has only been demonstrated to deliver a fraction of its theoretical capacity of 167 mAh/g (Refs. 9 to 12) at very low to modest discharge rates and severe capacity fading in systems with conventional Li-ion organic electrolytes containing $LiPF_6$ salts. Specific capacities reported in the literature range from 65 to 105 mAh/g at rates between C/50 and C/10 when the cathode is cycled between ~5.3 to 5.0 V to ~3.5 to 3.0 V at room temperature (Refs. 9 to 11, and 13). Although it is theorized that the likely culprit for the poor performance and high fade rate in the $LiCoPO_4$ cathode is the instability of the electrolyte (oxidation and decomposition) during high voltage operation and subsequent side reactions that inhibit lithium insertion and extraction (Refs. 9, 10, 13, and 14), operation at lower voltages would yield even lower specific capacities. Poor electronic conductivity of the active olivine material has also been reported (Refs. 9 and 13). The weightings of the chemistry options with respect to Rate Capability to C/5 are shown in Table 5.

Specific Energy

Quantitative specific energy projections from models were used to generate the pairwise comparisons for each chemistry with respect to every other chemistry. The specific energy was calculated using the projected specific capacity and voltage performance of each electrode after 3 years of focused development and the resultant performance of each of the pairs. The projected values for Specific Energy for each chemistry, when packaged in prismatic steel cases, and their weightings are given in Table 6.

TABLE 5.—WEIGHTING OF CHEMISTRY OPTIONS WITH RESPECT TO RATE CAPABILITY TO C/5

Anode	Cathode	Weight
Si-Composite	$Li(LiNMC)O_2$ (ETDP)	6.2
Si-Composite	$Li(Ni_{0.33}Mn_{0.33}Co_{0.33})O_2$	10.3
Li-metal	$Li(Ni_{0.33}Mn_{0.33}Co_{0.33})O_2$	16.5
Li-metal	$Li(LiNMC)O_2$ (ETDP)	10.6
Li-metal	$LiNiMn_2O_4$	31.3
Li-metal	$LiCoPO_4$	2.6
Li-metal	$(Li_2)S$	22.6

TABLE 6.—WEIGHTING OF CHEMISTRY OPTIONS WITH RESPECT TO SPECIFIC ENERGY

Anode	Cathode	Projected specific energy for a prismatic cell (Wh/kg)	Weight
Si-Composite	$Li(LiNMC)O_2$ (ETDP)	255	17.0
Si-Composite	$Li(Ni_{0.33}Mn_{0.33}Co_{0.33})O_2$	163	10.9
Li-metal	$Li(Ni_{0.33}Mn_{0.33}Co_{0.33})O_2$	199	13.3
Li-metal	$Li(LiNMC)O_2$ (ETDP)	243	16.2
Li-metal	$LiNiMn_2O_4$	180	12.0
Li-metal	$LiCoPO_4$	201	13.4
Li-metal	$(Li_2)S$	259	17.3

Storage and Calendar Life

The weightings for Storage and Calendar Life and their projected values are shown in Table 7. The clustering of the weights for this attribute is indicative of the large amount of uncertainty regarding the storage and calendar life of chemistries that will incorporate these advanced materials. Chemistries that have a 3-year life were projected to achieve this at a minimum—there were no known issues that could impact storage and calendar life.

Cells with Li-metal anodes were determined by the team to have a shorter storage and calendar life potential than cells with Si-based composite anodes. Li-metal rechargeable cells are built in a charged state. A continuous reaction between Li-metal in the charged state and the electrolyte and the ensuing impedance growth impacts their storage and calendar life. Alternate non-organic electrolytes, such as ionic liquids, have not been shown to alleviate this effect and may not be stable in a Li-metal rechargeable system. Li/S has a high self-discharge rate, and it is unknown if the lost capacity is reversible, so it is projected to have only about a 2-year life.

TABLE 7.—WEIGHTING OF CHEMISTRY OPTIONS
WITH RESPECT TO STORAGE AND CALENDAR LIFE

Anode	Cathode	Projected storage and calendar life (yr)	Weight
Si-Composite	$Li(LiNMC)O_2$ (ETDP)	>3	16.7
Si-Composite	$Li(Ni_{0.33}Mn_{0.33}Co_{0.33})O_2$	>3	16.7
Li-metal	$Li(Ni_{0.33}Mn_{0.33}Co_{0.33})O_2$	~2.5	13.9
Li-metal	$Li(LiNMC)O_2$ (ETDP)	~2.5	13.9
Li-metal	$LiNiMn_2O_4$	~2.5	13.9
Li-metal	$LiCoPO_4$	~2.5	13.9
Li-metal	$(Li_2)S$	~2	11.1

Energy Density

Energy density projections were made in a similar fashion to the specific energy projections discussed above. The projected values for Energy Density for each chemistry, when packaged in prismatic steel cases, and their weightings are given in Table 8. Although Li/S was weighted the highest for Specific Energy, it received the lowest weight for Energy Density.

TABLE 8.—WEIGHTING OF CHEMISTRY OPTIONS WITH RESPECT TO ENERGY DENSITY

Anode	Cathode	Projected energy density for a prismatic cell (Wh/L)	Weight
Si-Composite	$Li(LiNMC)O_2$ (ETDP)	646	19.5
Si-Composite	$Li(Ni_{0.33}Mn_{0.33}Co_{0.33})O_2$	436	13.2
Li-metal	$Li(Ni_{0.33}Mn_{0.33}Co_{0.33})O_2$	450	13.6
Li-metal	$Li(LiNMC)O_2$ (ETDP)	538	16.3
Li-metal	$LiNiMn_2O_4$	408	12.3
Li-metal	$LiCoPO_4$	428	13.0
Li-metal	$(Li_2)S$	403	12.2

Manufacturability

The team defined Manufacturability as the projected level of ease or difficulty associated with working with materials, scaling up batches of materials, manufacturing cells of practical capacity made from the components, and the projected adaptability of materials to large scale processing. All of the cathodes materials were determined to be similar enough to heritage Li-ion cathode materials to require similar manufacturing and cell processing techniques that would be compatible with or adaptable to currently existing manufacturing processes and equipment, with the exception of the sulfur cathode. Most of the Li-metal chemistries were determined to be highly manufacturable, based on the established knowledge base in manufacturing Li primary (nonrechargeable) cells. Li/S was again the exception due to the unknowns associated with manufacturing large capacity sulfur cathodes and incorporating them into a cell.

It was determined that Si-based composite anodes would be more difficult to manufacture than Li-metal anodes. There are currently no known US manufacturers of Si-based composite anodes to make up a domestic knowledge base for large scale processing of these materials. Chemical vapor deposition is a common technique used in laboratories to deposit (or plate) Si on a substrate. It is unknown if this process is scalable to manufacture large quantities and sizes of anode materials. Weights for Manufacturability are shown in Table 9.

TABLE 9.—WEIGHTING OF CHEMISTRY OPTIONS WITH RESPECT TO MANUFACTURABILITY

Anode	Cathode	Weight
Si-Composite	$Li(LiNMC)O_2$ (ETDP)	7.8
Si-Composite	$Li(Ni_{0.33}Mn_{0.33}Co_{0.33})O_2$	7.8
Li-metal	$Li(Ni_{0.33}Mn_{0.33}Co_{0.33})O_2$	20.5
Li-metal	$Li(LiNMC)O_2$ (ETDP)	20.5
Li-metal	$LiNiMn_2O_4$	20.5
Li-metal	$LiCoPO_4$	20.5
Li-metal	$(Li_2)S$	2.5

Schedule

Chemistries with the $Li(Ni_{0.33}Mn_{0.33}Co_{0.33})O_2$ cathode fared well for schedule since it is a commercially available material that would only require minor tweaks to optimize it for the specific chemistry. Although the anode paired with the $Li(Ni_{0.33}Mn_{0.33}Co_{0.33})O_2$ cathode requires development, utilizing a cathode with proven performance greatly reduces schedule risks.

The development of a Si-based composite anode has a lower schedule risk than developing a stable Li-metal rechargeable chemistry. The development of compatible electrolytes and effective coatings are critical to improve the safety of chemistries containing a Li-metal anode and may pose significant schedule issues. Therefore, the Si-composite/$Li(LiNMC)O_2$ cathode weighting is higher than the $Li(Ni_{0.33}Mn_{0.33}Co_{0.33})O_2$ cathode paired with a Li-metal anode. Results of the weighting of chemistries with respect to Schedule are shown in Table 10.

TABLE 10.—WEIGHTING OF CHEMISTRY OPTIONS WITH RESPECT TO SCHEDULE

Anode	Cathode	Weight
Si-Composite	$Li(LiNMC)O_2$ (ETDP)	16.8
Si-Composite	$Li(Ni_{0.33}Mn_{0.33}Co_{0.33})O_2$	38.0
Li-metal	$Li(Ni_{0.33}Mn_{0.33}Co_{0.33})O_2$	15.8
Li-metal	$Li(LiNMC)O_2$ (ETDP)	10.8
Li-metal	$LiNiMn_2O_4$	8.6
Li-metal	$LiCoPO_4$	4.6
Li-metal	$(Li_2)S$	5.4

Cost to TRL 6

The Cost to TRL 6 attribute considered the cost to develop the technology to TRL 6, including costs attributed to costly manufacturing processes or processes that cannot be automated. In the absence of true costs, this factor was judged qualitatively by performing pairwise comparisons of the relative cost to develop each chemistry with respect to every other chemistry. The results are shown in Table 11.

TABLE 11.—WEIGHTING OF CHEMISTRY OPTIONS WITH RESPECT TO COST TO TRL 6

Anode	Cathode	Weight
Si-Composite	$Li(LiNMC)O_2$ (ETDP)	24.1
Si-Composite	$Li(Ni_{0.33}Mn_{0.33}Co_{0.33})O_2$	33.3
Li-metal	$Li(Ni_{0.33}Mn_{0.33}Co_{0.33})O_2$	14.0
Li-metal	$Li(LiNMC)O_2$ (ETDP)	7.6
Li-metal	$LiNiMn_2O_4$	10.7
Li-metal	$LiCoPO_4$	3.5
Li-metal	$(Li_2)S$	6.8

Cycle Life

The cycle life of each chemistry after 3 years of development was projected. Cycle life at C/5 to 100 percent DOD and to 80 percent of initial capacity was used as a benchmark to qualify cycle life. Some general observations regarding potential cycle life of the chemistries were noted. Si-based composite chemistries will be anode limited and were projected to achieve approximately 200 cycles under the given conditions. With the exception of the spinel and olivine compositions and Li/S, the Li-metal chemistries would also be anode limited to achieve the maximum cycles Li-metal is capable of delivering. This was approximated at 300 cycles under the conditions listed above. For the other Li-metal chemistries, the cycle life of the cathode was the determining factor. Operation at high voltages is expected to adversely impact the cycle life of the spinel and olivine cathodes (Ref. 15). The projected cycle lives of the chemistries and their weights are shown in Table 12.

TABLE 12.—WEIGHTING OF CHEMISTRY OPTIONS WITH RESPECT TO CYCLE LIFE

Anode	Cathode	Projected cycle life	Weight
Si-Composite	$Li(LiNMC)O_2$ (ETDP)	200	14.3
Si-Composite	$Li(Ni_{0.33}Mn_{0.33}Co_{0.33})O_2$	200	14.3
Li-metal	$Li(Ni_{0.33}Mn_{0.33}Co_{0.33})O_2$	300	21.4
Li-metal	$Li(LiNMC)O_2$ (ETDP)	300	21.4
Li-metal	$LiNiMn_2O_4$	200	14.3
Li-metal	$LiCoPO_4$	100	7.1
Li-metal	$(Li_2)S$	100	7.1

Rate Capability to C/2

The logic to project rate capability at C/2 paralleled the logic for C/5 performance. However, since the $LiNiMn_2O_4$ spinel oxide cathode can deliver superior performance at higher rates, it was weighted more heavily at C/2. The performance of Li/S degrades much more rapidly at C/2, so the $Li/Li(Ni_{0.33}Mn_{0.33}Co_{0.33})O_2$ chemistry is weighted higher here. The weightings of the chemistry options with respect to Rate Capability to C/2 are shown in Table 13.

TABLE 13.—WEIGHTING OF CHEMISTRY OPTIONS WITH RESPECT TO RATE CAPABILITY TO C/2

Anode	Cathode	Weight
Si-Composite	$Li(LiNMC)O_2$ (ETDP)	7.0
Si-Composite	$Li(Ni_{0.33}Mn_{0.33}Co_{0.33})O_2$	8.8
Li-metal	$Li(Ni_{0.33}Mn_{0.33}Co_{0.33})O_2$	17.4
Li-metal	$Li(LiNMC)O_2$ (ETDP)	11.1
Li-metal	$LiNiMn_2O_4$	39.3
Li-metal	$LiCoPO_4$	2.2
Li-metal	$(Li_2)S$	14.2

Results

An analysis of the results of the chemistry rankings with respect to each attribute reveals that Si-based composite chemistries were highly favored with respect to four of the ten attributes: Safety, Storage and Calendar Life, Schedule, and Cost to TRL 6. Li-metal chemistries tended to be weighted highest with respect to three of the ten attributes: Rate Capability to C/5, Manufacturability, and Rate Capability to C/2. Both anode types received a range of weightings in the categories of Specific Energy, Energy Density, and Cycle Life, depending upon the cathode they were paired with.

Table 14 shows the final overall weightings of all the seven chemistries. The dominance of the weightings of the Si-based composite chemistries with respect to Safety, the most important attribute (as shown in Table 3), tilted the final weightings in favor of the Si-based composite chemistries. The Si-based composite/Li($Ni_{0.33}Mn_{0.33}Co_{0.33}$)$O_2$ chemistry received the highest overall ranking. Since Li($Ni_{0.33}Mn_{0.33}Co_{0.33}$)$O_2$ is a commercial cathode, the chemistry was a clear preference in categories such as Schedule and Cost to TRL 6. This cathode also contributed to the chemistry's higher weightings over the Si-based composite/Li(LiNMC)O_2 chemistry for factors such as Safety and Rate Capability to C/5, the two most important attributes, in addition to Energy Density and Rate Capability to C/2. The sole attribute in which the Si-based composite/Li(LiNMC)O_2 chemistry outperformed the Li($Ni_{0.33}Mn_{0.33}Co_{0.33}$)$O_2$ counterpart is Specific Energy. Specific Energy is the third most important attribute, and is virtually tied with the second most important attribute.

TABLE 14.—FINAL WEIGHTING OF CHEMISTRY OPTIONS

Anode	Cathode	Final weight
Si-Composite	Li($Ni_{0.33}Mn_{0.33}Co_{0.33}$)$O_2$	20.2
Si-Composite	Li(LiNMC)O_2 (ETDP)	17.0
Li-metal	$LiNiMn_2O_4$	15.3
Li-metal	Li($Ni_{0.33}Mn_{0.33}Co_{0.33}$)$O_2$	13.9
Li-metal	Li(LiNMC)O_2 (ETDP)	13.1
Li-metal	(Li_2)S	11.5
Li-metal	$LiCoPO_4$	9.1

Given the results of the rigorous ranking process, the team determined the best course of action to pursue to develop the chemistry that could deliver the best combination of safety and performance in the available timeframe. Given that the Li(LiNMC)O_2 ETDP cathode is already under development in the project and the Li($Ni_{0.33}Mn_{0.33}Co_{0.33}$)$O_2$ cathode is a commercial material that would only need minor tweaking to optimize its performance in conjunction with other cell components, a decision was made to continue development of the Li(LiNMC)O_2 ETDP cathode to pursue higher potential gains in specific energy, and to hold the Li($Ni_{0.33}Mn_{0.33}Co_{0.33}$)$O_2$ chemistry in reserve as a back-up option.

Key Performance Parameters

A set of Key Performance Parameters (KPPs) were established for the HE and UHE cells through a combination of customer requirements definition and results of the feasibility study, considering the chemistry chosen and expert opinions on what technology advancements could reasonably deliver with the available resources on the required schedule. The UHE cell development will enable a battery system that can operationally deliver 220 Wh/kg at the beginning of life and can provide 200 cycles to 80 percent of its original capacity. The UHE cell development adds advanced anodes to the existing component development portfolio for HE cells to enable much higher cell-level specific energy, on the order of a 120 percent increase over SOA.

The KPP goals are shown in Table 15. The first column contains customer needs, which have been tempered based on the capability of the technology that was chosen to address the full range of requirements. SOA values are given in the third column of Table 15. Specific energy and energy density performance are interrelated with rate capability and other operating conditions. A baseline set of conditions was specified at which specific energy and energy density performance of the SOA cells and battery would be stated in order to compare advanced developments to existing technology. These conditions are operation at C/10 and 0 °C to 100 percent depth-of-discharge (DOD), defined as a discharge to 3.0 V.

SOA aerospace Li-ion cell technology was determined to be the cells that make up the 2003 MER rover batteries. Since there is no direct comparison for SOA batteries operating in a Lunar environment, it was determined that the MER rover batteries represent a closer case study than Li-ion cells operating on an Earth-orbiting satellite. These batteries are at TRL 9 since they have achieved operation under actual mission conditions, they are operating under a similar thermal environment as the Lunar batteries are expected to operate under (with thermal controls) and they will likely operate under similar cyclical profiles, i.e., providing primary power for several hours on a single discharge for missions lifetimes of several months.

TABLE 15.—KEY PERFORMANCE PARAMETERS

Customer need	Performance parameter	SOA	Current value	Threshold value	Goal
Safe, reliable operation	No fire or flame	Instrumentation/controllers used to prevent unsafe conditions. There is no nonflammable electrolyte in SOA	Preliminary results indicate a moderate reduction in the performance with flame retardants and nonflammable electrolytes	Benign cell venting without fire or flame and reduce the likelihood and severity of a fire in the event o f a thermal runaway	Tolerant to electrical and thermal abuse such as over-temperature, overcharge, reversal, and external short circuit with no fire or flame
Specific energy Lander: 150 to 210 Wh/kg 10 cycles Rover: 150 to 200 Wh/kg EVA: 180 to 230 Wh/kg 100 cycles	Battery-level specific energy	90 Wh/kg at C/10 and 30 °C 83 Wh/kg at C/10 and 0 °C (MER rovers)	130 Wh/kg at C/10 and 30 °C 120 Wh/kg at C/10 and 0 °C	135 Wh/kg at C/10 and 0 °C HE 150 Wh/kg at C/10 and 0 °C UHE	150 Wh/kg at C/10 and 0 °C HE 220 Wh/kg at C/10 and 0 °C UHE
	Cell-level specific energy	130 Wh/kg at C/10 and 30 °C 118 Wh/kg at C/10 and 0 °C	150 Wh/kg at C/10 and 0 °C	165 Wh/kg at C/10 and 0 °C HE 180 Wh/kg at C/10 and 0 °C UHE	180 Wh/kg at C/10 and 0 °C HE 260 Wh/kg at C/10 and 0 °C UHE
	Cathode-level specific capacity $Li(Li,NiMn)O_2$	140 to 150 mAh/g typical	$Li(Li_{0.17}Ni_{0.25}Mn_{0.58})O_2$: 240 mAh/g at C/10 and 25 °C $Li(Li_{0.2}Ni_{0.13}Mn_{0.54}Co_{0.13})O_2$: 250 mAh/g at C/10 and 25 °C 200 mAh/g at C/10 and 0 °C	260 mAh/g at C/10 and 0 °C	280 m Ah/g at C/10 and 0 °C
	Anode-level specific capacity	320 mAh/g (MCMB)	320 mAh/g (MCMB) 450 mAh/g Si composite	600 mAh/g at C/10 and 0 °C (with Si composite)	1000 m Ah/g at C/10 0 °C (with Si composite)
Energy density Lander: 311 Wh/L Rover: TBD EVA: 400 to 700 Wh/L	Battery-level energy density	250 Wh/L	N/A	270 Wh/L HE 360 Wh/L UHE	320 Wh/L HE 420 Wh/L UHE
	Cell-level energy density	320 Wh/L	N/A	385 Wh/L HE 460 Wh/L UHE	390 Wh/L HE 530 Wh/L UHE
Operating environment 0 to 30 °C, vacuum	Operating temperature	–20 to 40 °C	–50 to 40 °C	0 to 30 °C	0 to 30 °C

Assumes prismatic cell packaging for threshold values. Goal values include lightweight battery packaging. Battery values are assumed at 100 percent DOD, discharged at C/10 to 3.0 V/cell, and at 0 °C operating conditions.

The Current Value column in Table 15 gives values that correspond to the capability of the advanced technology that has been developed to date under the ETDP project (cathodes and electrolytes) or literature values (anodes). Cell and battery-level values in this column were projected from the performance capability of the advanced components. The Threshold values are the minimum acceptable performance criteria for success. The Goal values are those that represent full success in meeting the project goals.

It is worthwhile to note that the UHE cell development is a higher risk approach than the HE cells, due to the combination of potential safety concerns with the use of pure Li metal anodes and Li-based anodes, low TRL for the anode materials, and the departure from traditional lithium-ion anode materials which will demand different types of processing and manufacturing approaches. Although the HE cell development is primarily targeted for Mobility Systems, due to the high risk nature of the UHE cell development, the HE cell will also serve as a backup technology for Altair and EVA.

Conclusions

The feasibility study resulted in the determination that Si-based composite/$Li(Ni_{0.33}Mn_{0.33}Co_{0.33})O_2$ and Si-based composite/$Li(LiNMC)O_2$ chemistries can provide the best combination of safety, specific energy, energy density, and likelihood of success in the available timeframe. Given these results, the ETDP Energy Storage Project made a decision to continue to develop the ETDP cathode, the $Li(LiNMC)O_2$, and to reserve the option to incorporate $Li(Ni_{0.33}Mn_{0.33}Co_{0.33})O_2$, optimized for the specific chemistry, as a back-up cathode. This strategy reduces the risk associated with cathode development at virtually no cost and provides the opportunity for the project to continue to develop the cathode with the potential to deliver much higher specific energy.

The project will also begin to aggressively develop Si-based composite anode materials. A safe, high specific capacity anode is critical to achieving the specific energy goals for the project. Si-based composite materials require development to address volume expansion to improve their mechanical and cycling stability and to reduce the first cycle irreversible capacity loss. These challenges were deemed to be addressable within the timeframe available.

Compatible electrolytes, separators, and safety components will also be developed. Electrolytes that are stable at high voltages, non-flammable electrolytes, and those that incorporate flame-retardant additives will be developed to improve the voltage stability and the safety of the chemistry. Shut-down separators, safer separators and safety components that are either incorporated into the electrochemical components of the cell or consist of functional, internal cell components, will also be developed and investigated.

In general, Li-metal chemistry options suffered from significant safety issues that need to be resolved. Appropriate electrolytes and coatings would have to be developed to overcome these issues. The significant challenges to achieving these goals led to a lack of confidence that the materials would be mature enough within the available timeframe. Li/S offers impressive specific energy performance and may be considered for development for far term missions or for future upgrades to current technology solutions.

Appendix A.—Abbreviations

°C	degrees Celsius
Ah	Ampere hours
Co	cobalt
DOD	depth of discharge
ETDP	Exploration Technology Development Program
EVA	extravehicular activities
HE	high energy
hr	hour(s)
KPP	Key Performance Parameter
Li	lithium
Li-ion	lithium-ion
LiNMC	lithium nickel manganese cobalt
Li/S	lithium sulfur
LSS	Lunar Surface Systems
LTO	lithium titanate
mAh/g	milliampere hours per gram
MCMB	mesocarbon microbeads
MER	Mars Exploration Rover
Mn	manganese
Ni	nickel
O	oxygen
P	phosphate
S	sulfur
Si-comp	silicon composite
SOA	state of the art
TRL	Technology Readiness Level
UHE	ultra high energy
V	Volts
Wh/kg	Watt-hours per kilogram

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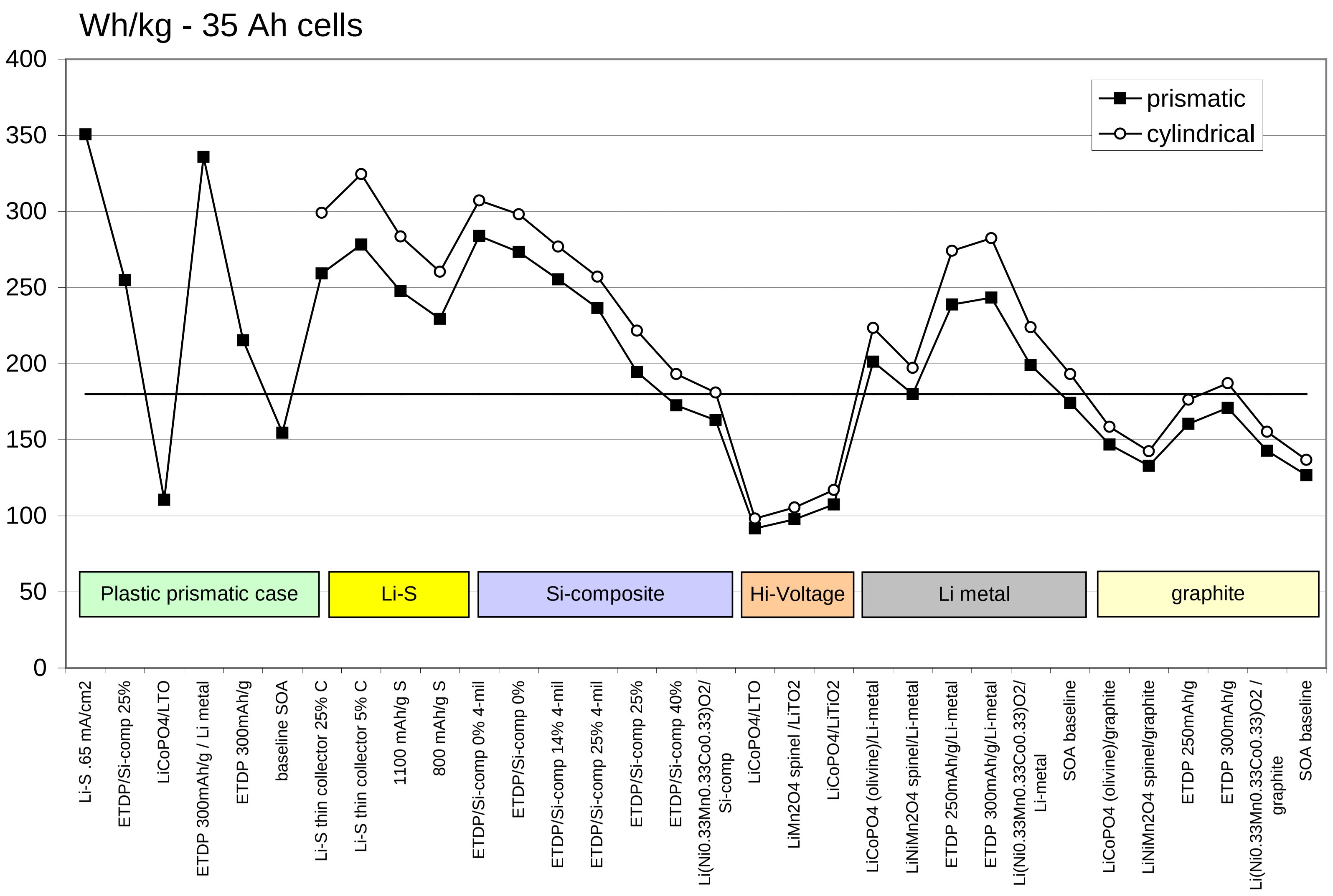

Figure 1.—Projected specific energy for chemistry options.

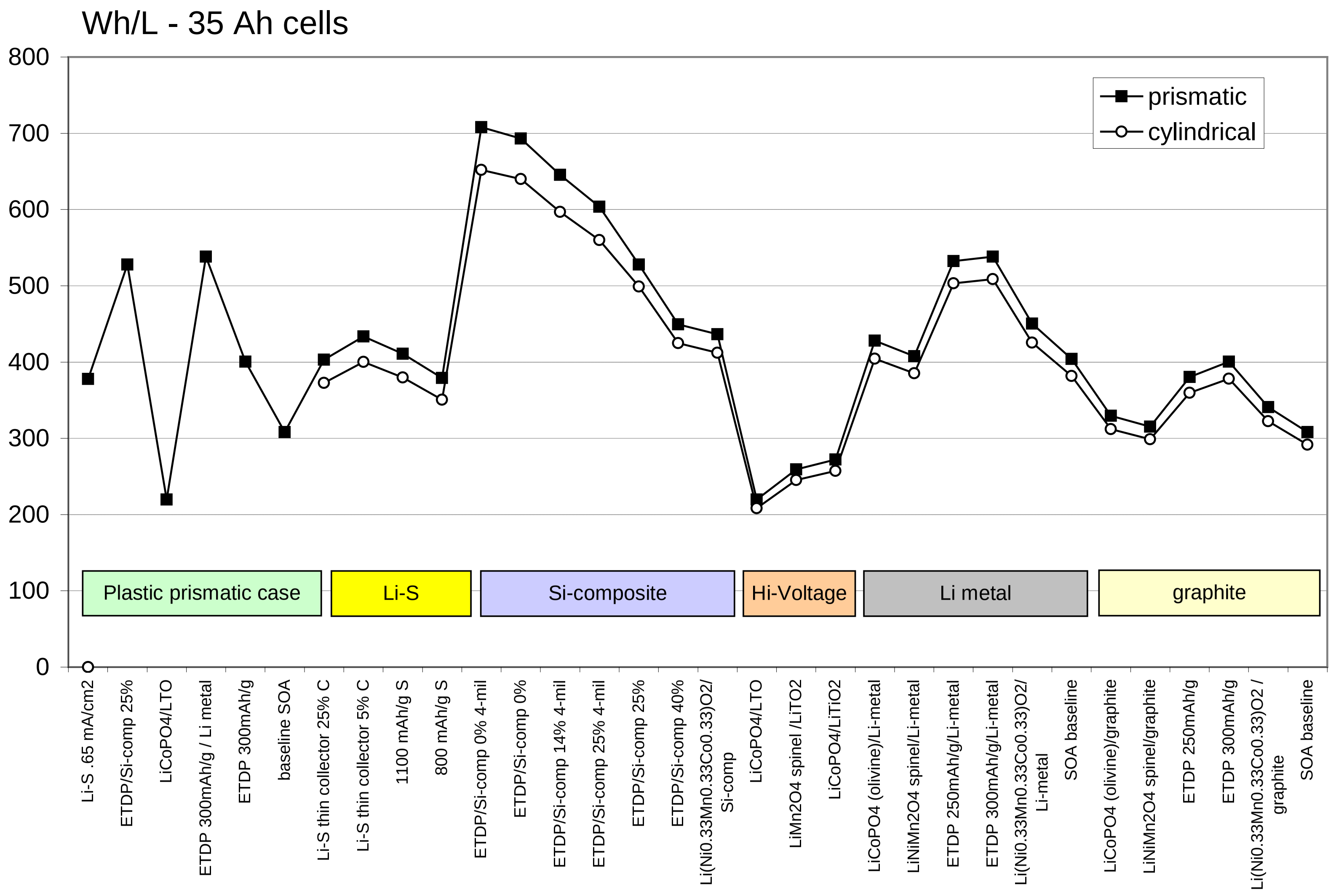

Figure 2.—Projected energy density for chemistry options.

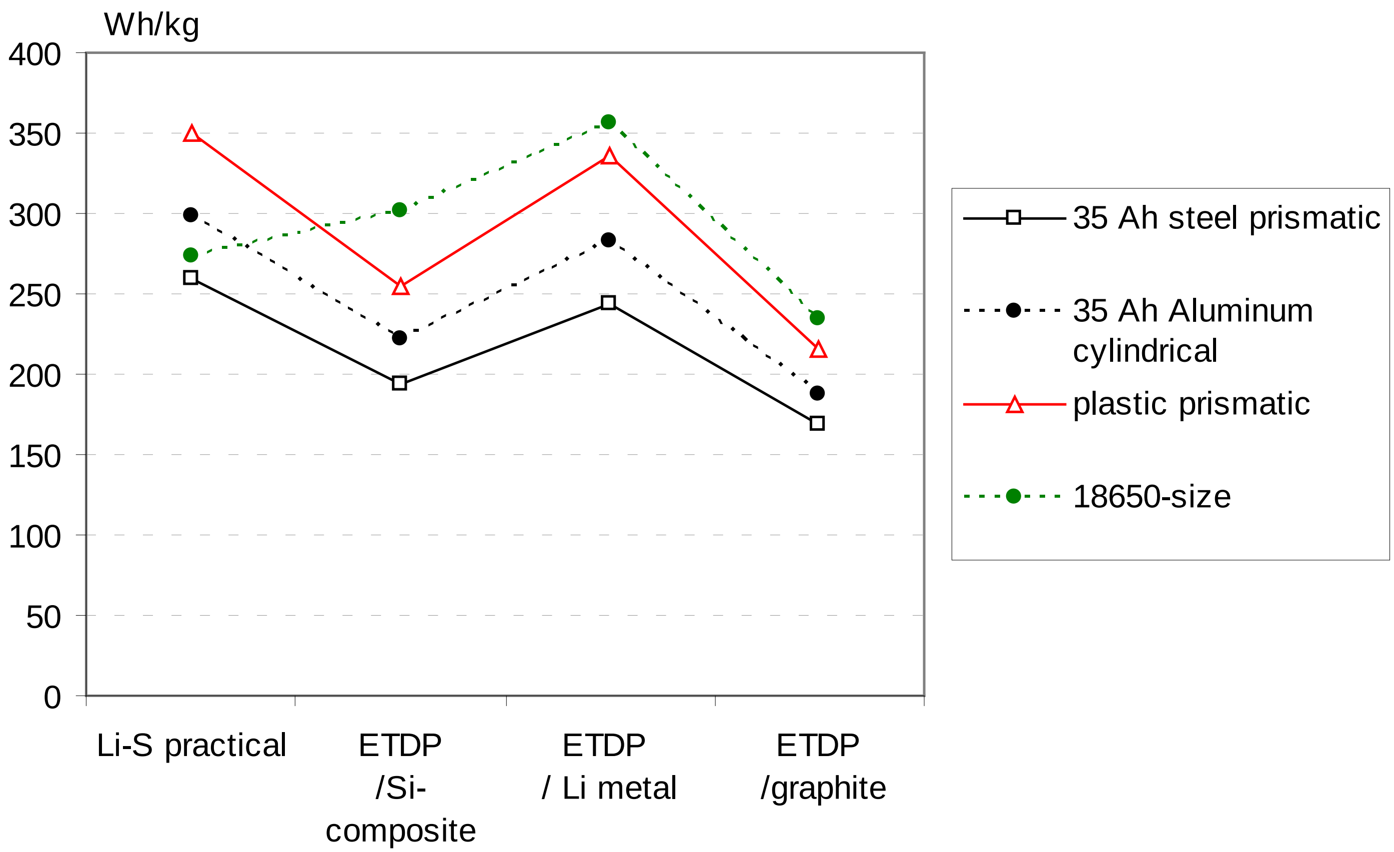

Figure 3.—Cell design effects on specific energy.

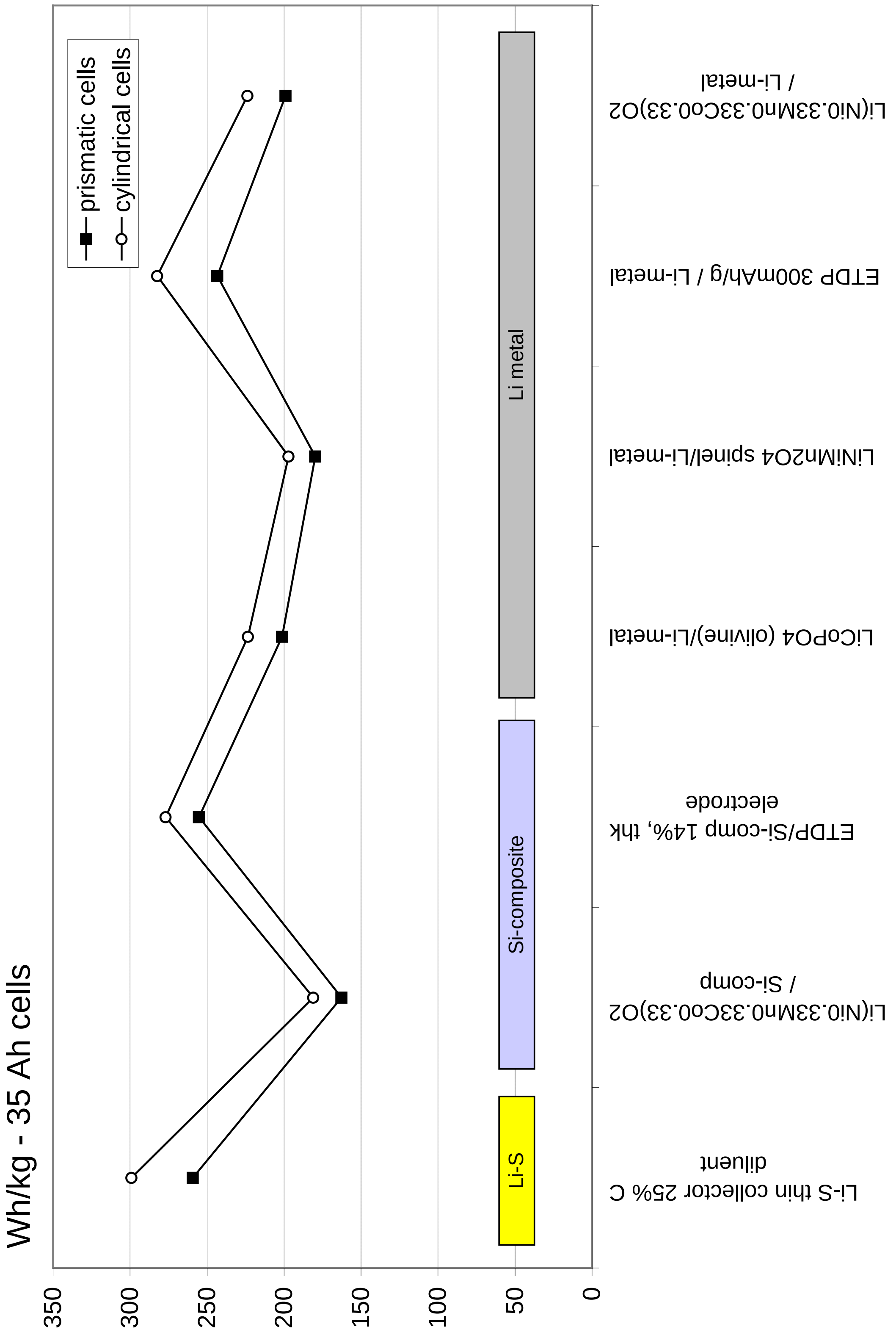

Figure 4.—Projected specific energy for final options.

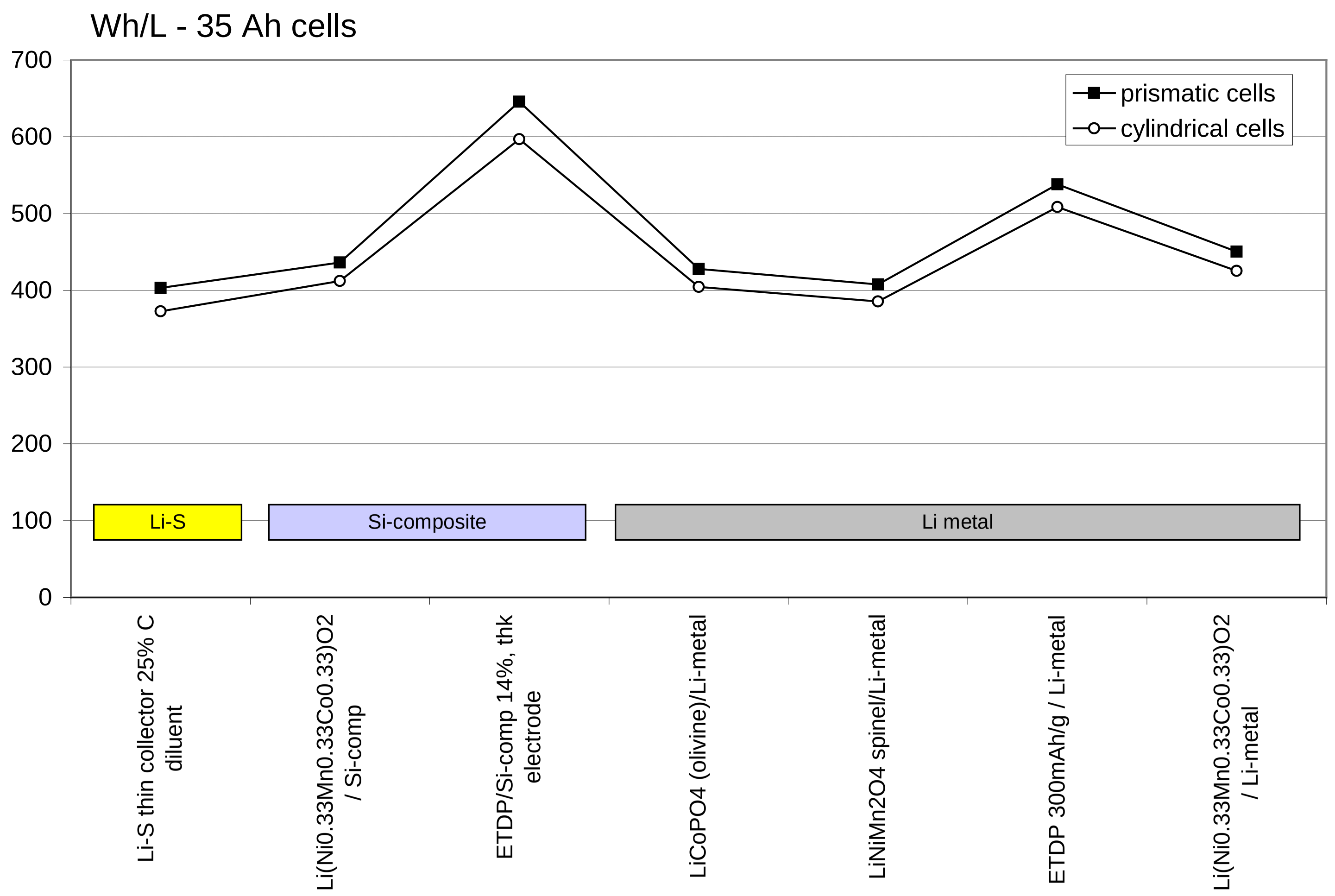

Figure 5.—Projected Energy Density for Final Options

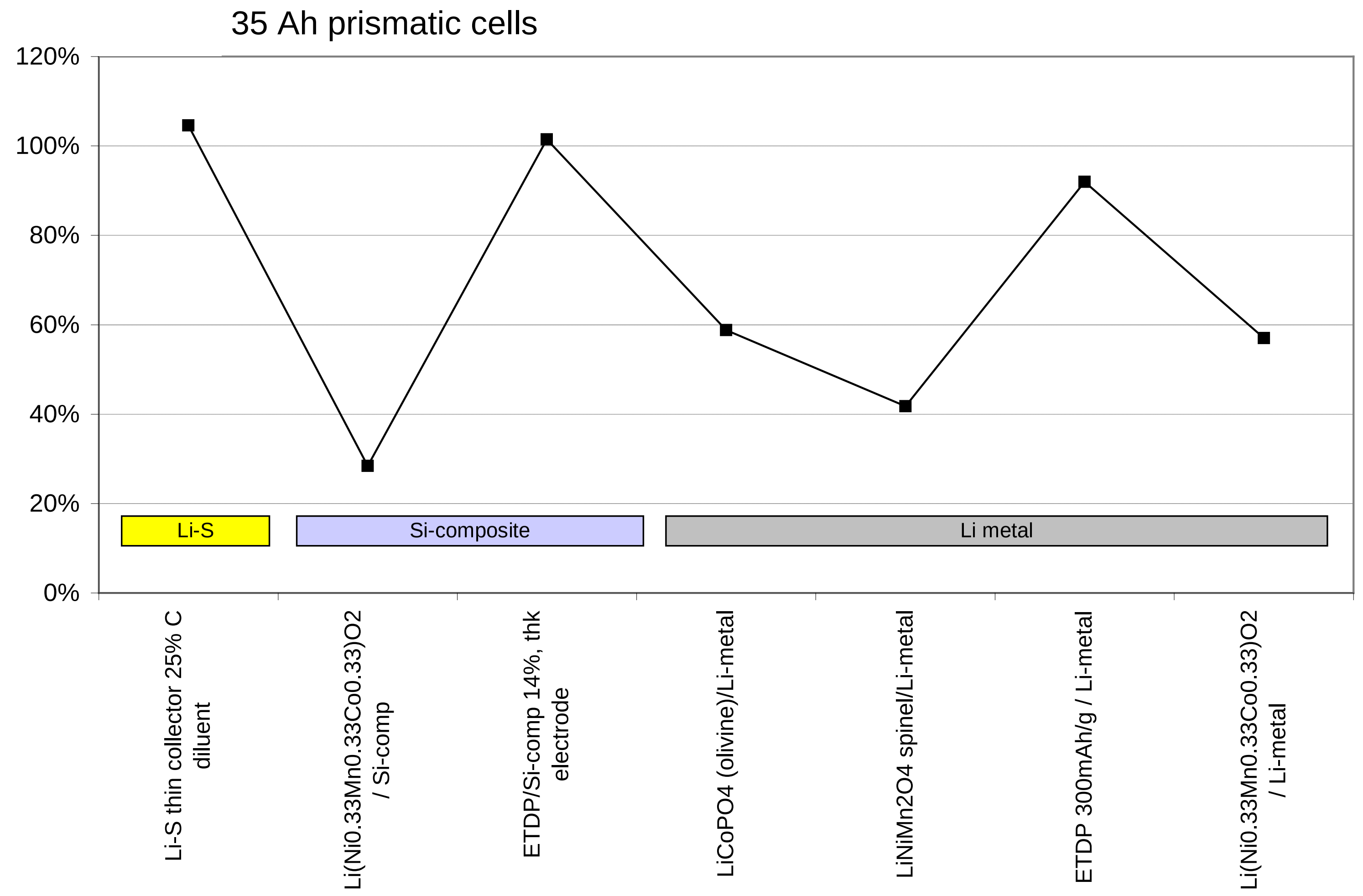

Figure 6.—Percent gain in specific energy of final options over Li-ion SOA cells.

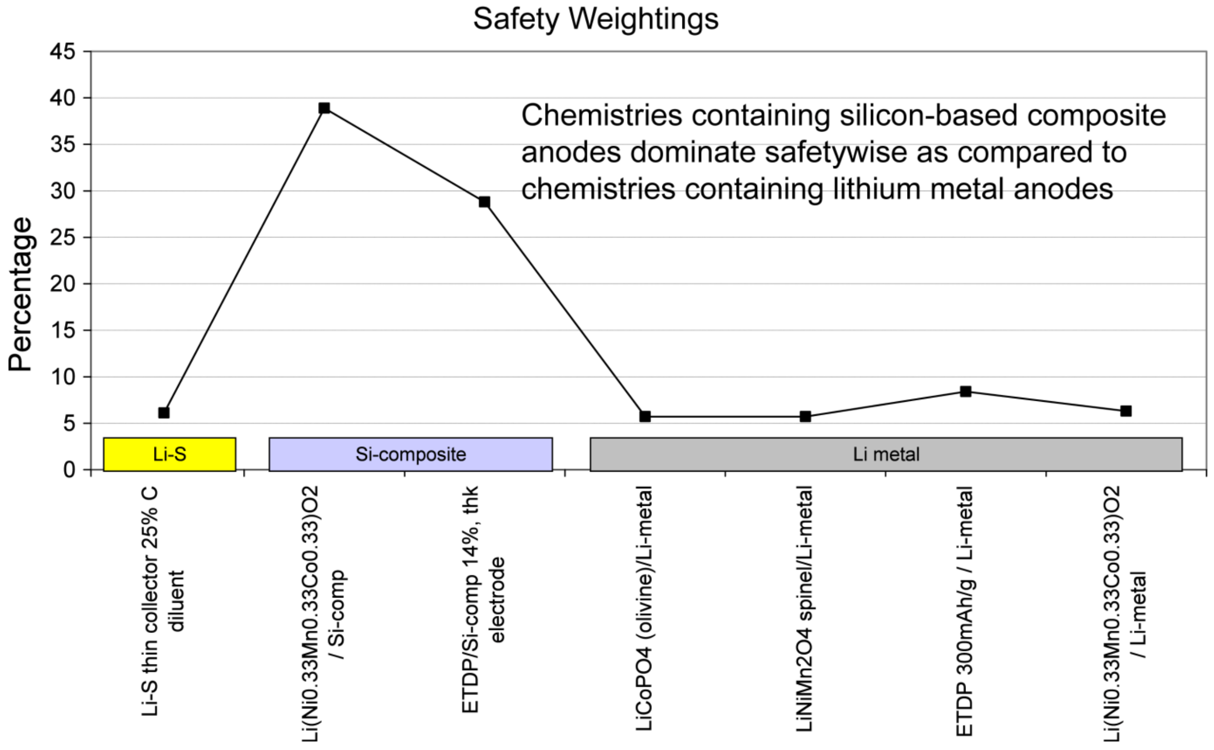

Figure 7.—Safety weightings for each chemistry.
(Sum of individual weights totals 100.)

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13. SUPPLEMENTARY NOTES

14. ABSTRACT

NASAs Exploration Technology Development Program (ETDP) Energy Storage Project conducted an advanced lithium-based battery chemistry feasibility study to determine the best advanced chemistry to develop for the Altair Lunar Lander and the Extravehicular Activities (EVA) advanced Lunar surface spacesuit. These customers require safe, reliable batteries with extremely high specific energy as compared to state-of-the-art. The specific energy goals for the development project are 220 watt-hours per kilogram (Wh/kg) delivered at the battery-level at 0 degrees Celsius (°C) at a C/10 discharge rate. Continuous discharge rates between C/5 and C/2, operation between 0 and 30 °C and 200 cycles are targeted. Electrode materials that were considered include layered metal oxides, spinel oxides, and olivine-type cathode materials, and lithium metal, lithium alloy, and silicon-based composite anode materials. Advanced cell chemistry options were evaluated with respect to multiple quantitative and qualitative attributes while considering their projected performance at the end of the available development timeframe. Following a rigorous ranking process, a chemistry that combines a lithiated nickel manganese cobalt oxide $Li(LiNMC)O_2$ cathode with a silicon-based composite anode was selected as the technology that can potentially offer the best combination of safety, specific energy, energy density, and likelihood of success.

15. SUBJECT TERMS
Lithium batteries; Storage batteries; Electric batteries; Electrochemical cells; Energy storage; Space missions; Spacecraft power systems; Electric power systems

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