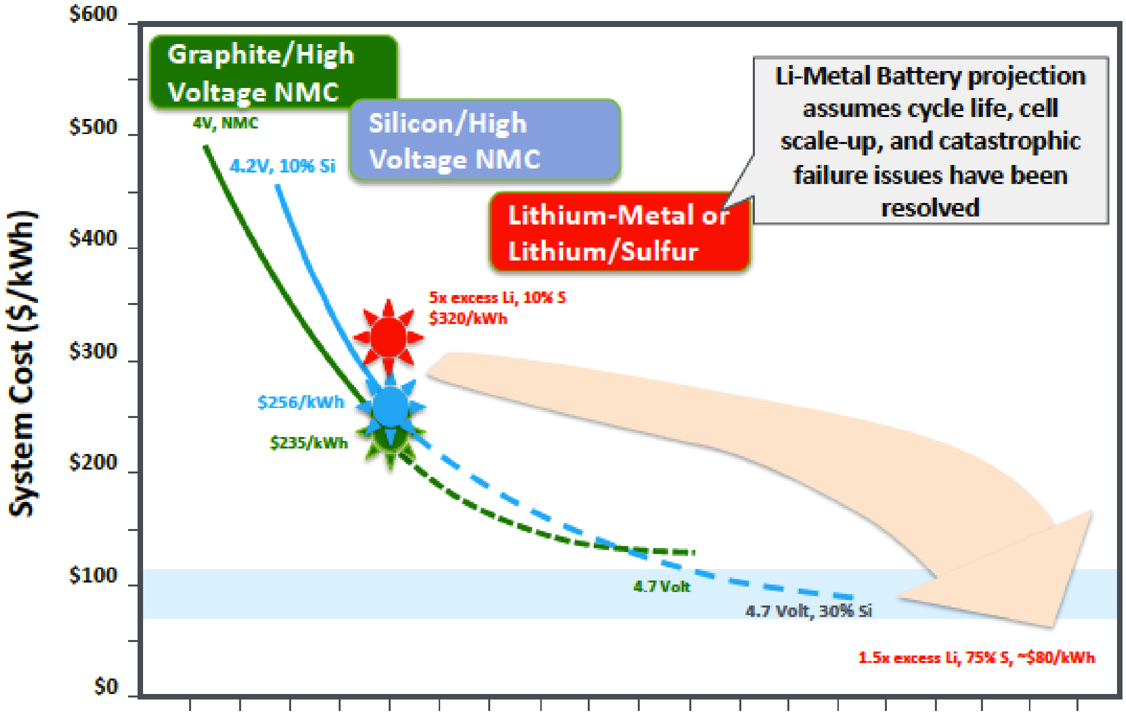
DRIVING RESEARCH AND INNOVATION FOR VEHICLE EFFICIENCY AND ENERGY SUSTAINABILITY

Electrochemical Energy Storage Technical Team Roadmap

September 2017



The potential Electric vehicle battery cost decrease over time, assuming successful research results in three technology areas

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U.S. DRIVE Electrochemical Energy Storage R&D Roadmap

Introduction

This U.S. DRIVE electrochemical energy storage roadmap describes ongoing and planned efforts to develop electrochemical energy storage technologies for electric drive vehicles, primarily plug-in electric vehicles (PEVs) and 12V start/stop (S/S) micro-hybrid batteries. Note that PEVs include both pure electric vehicles (EV) and plug-in hybrid electric vehicles (PHEV) that contain an internal combustion engine to extend range. The energy storage activity comprises a number of research areas (e.g., advanced battery material R&D and advanced battery cell R&D) with the goal of developing energy storage devices for more fuel-efficient light duty vehicles that can reduce U.S. dependence on petroleum without sacrificing performance.

One of the ultimate goals of this research, and currently a strong trend in vehicle electrification, is the EV which should provide the full driving performance, convenience, and price of an internal combustion engine (ICE) vehicle. The advantages of EVs include very high efficiency compared to other vehicle types, a simplified drive train, and the increased flexibility of the primary energy source. Electricity used to charge the EV can be generated from coal, natural gas, wind turbines, hydroelectric, solar energy, nuclear, or any other resource. Another current focus is the 12V start/stop (S/S) micro-hybrid architecture, in which the engine is shut down whenever the vehicle stops. Vehicles with S/S functionality are being deployed worldwide to address emissions concerns. The 12V battery provides power for auxiliary equipment (like the radio and air conditioning) and then restarts the engine when the vehicle moves. Current 12V start/stop batteries are typically lead-acid and have poor life. Several automakers are considering 48V systems and U.S. DRIVE may, in the future, conduct research into batteries for this application.

Over the past seven years, EVs have become much more commercially viable, with battery costs dropping almost 80% since 2010, and with the introduction of vehicles like the Chevy Bolt EV with an EPA-estimated 238-mile range and a retail price of $37,495. But better and less expensive energy storage systems are still needed to expand the commercial markets for EVs, which currently sell at ~1% of new vehicle sales. Lower-cost batteries with higher energy density, higher power (including the ability to accept extreme fast charging [XFC]), and better low-temperature operation, are needed to give EVs the same all-weather performance and “refueling” convenience as ICE vehicles. Lithium (Li)-based batteries offer the best chance to meet the requirements and are the primary focus of U.S. DRIVE.

The U.S. DRIVE Electrochemical Energy Storage Tech Team has been tasked with providing input to DOE on its suite of energy storage R&D activities. The members of the tech team include: General Motors, Ford Motor Company, Fiat-Chrysler Automotive; and the Electric Power Research Institute (EPRI). To facilitate exchange of information between the team members and DOE program managers, periodic meetings are held at which specific research topics are presented to the Tech Team for discussion. Sample topics include advanced manufacturing R&D, extreme fast charge, silicon anode R&D needs, etc.

I. Goals

Table 1 and 2 show a subset of the targets for EV and 12V start/stop micro hybrid batteries that have been developed by U.S. DRIVE. Extreme fast charge cell targets are shown in Section III.2.c.

Table 1. Subset of EV for batteries and cells. Red shading = current commercial cells are not meeting the goal. Cost and low temperature performance are critical requirements.

Energy Storage Goals	System Level	Cell Level
Characteristic		
Cost @ 100k units/year (kWh = useable energy)	$100/kWh	$75/kWh
Peak specific discharge power (30s)	470 W/kg	700 W/kg
Peak specific regen power (10s)	200 W/kg	300 W/kg
Useable specific energy (C/3)	235 Wh/kg	350 Wh/kg
Usable energy density (C/3)	500 Wh/l	750 Wh/l
Calendar life	15 years	15 years
Deep discharge cycle life	1000 cycles	1000 cycles
Low temperature performance	>70% useable energy @C/3 discharge at -20°C	>70% useable energy @C/3 discharge at -20°C

Table 2. Subset of targets for 12V start/stop micro-hybrid batteries. Red shading = current commercial cells are not meeting the goal. Yellow shading = current cells are almost meeting they goal. Cost and cold cranking are critical requirements.

Energy Storage Goals	Under hood	Not under hood
Characteristic		
Maximum selling price	$220	$180
Discharge pulse (1s)	6 kW	
Cold cranking power, (-30°C)	6 kW for 0.5s followed by three 4 kW/4s pulses	
Available energy	360 Wh	
Peak recharge rate (10s)	2.2 kW	
Sustained recharge rate	750 W	
Cycle life	450 k	
Calendar life	15 years at 45°C	15 years at 30°C
Maximum weight	10 kg	
Maximum volume	7 liters	

II. Challenges and Barriers

There has been significant progress in developing Li-ion EV batteries in terms of their weight, volume, life, and even cost. The major remaining challenges to further commercializing batteries for EVs and 12V start/stop micro-hybrid batteries are as follows:

A. Cost. The current cost of high-energy Li-ion batteries is approximately $200 - $300/kWh (usable energy), a factor of two- three times too high. Cost of Li-ion based 12V micro-hybrid batteries (which offer significantly better life) is approximately 50% too high compared to their lead acid counterparts. The main cost drivers are the high cost of raw materials and materials processing, the cost of cell and module packaging, and manufacturing costs.

- **Beyond Li-ion:** Cells containing Li metal anodes
- **Next-gen Li-ion:** Cells containing an alloy anode, usually Silicon based, and or a high voltage (>4.5 V) cathode
- **Enhanced Li-ion:** Cells with today's materials (graphite anode/ transition metal oxide cathode), but with features like XFC, low T performance, and improved abuse tolerance

B. Performance. Historically, higher energy density was needed to reduce the weight and volume of PEV batteries, but weight and volume have been mainly addressed. Higher energy cells are still one way to reduce costs, however, cell chemistries that provide higher energy have life and performance issues. Also, existing chemistries (graphite anodes paired with transition metal oxide cathodes) need improvement in XFC and low temperature performance to match gas-powered vehicles' performance and customer convenience. The main performance issue with Li-ion 12V start/stop batteries is the challenging "cold start" requirement at -30°C.

C. Life. The life issue for mature Li-ion technologies has mainly been addressed. However, next gen (e.g., cells containing an alloy anode, usually silicon based, and or a high voltage (>4.5 volts) cathode) and beyond Li-ion (BLI, cells containing Li metal anodes) technologies still suffer major cycle and calendar life issues. The life of Li-ion based 12V start/stop micro-hybrid batteries is relatively good, however it is often the case that enhancing cold crank performance shortens high temperature life.

D. Abuse Tolerance. Current Li-ion automotive batteries are being used in a safe manner. Thus, although Li-ion is not intrinsically tolerant to abusive conditions (neither is gasoline), it can be engineered in a commercially acceptable product. The characteristics of next-gen and BLI chemistries to abusive conditions are largely unknown. However, Li-metal based batteries have a long history of problematic dendrite growth which leads to internal shorts and thermal runaway.

E. Recycling and Sustainability. Currently, automotive OEMs pay a relatively large cost (5-15% of the battery cost) to recycle end of life PEV batteries. The various chemistries used in Li-ion cells results in variable backend value. Alternatively, not recycling Li-ion batteries may lead to a shortage of key materials (lithium, cobalt, and nickel) that are vital to the technology. Finding ways to decrease the cost of recycling could thus significantly reduce the life cycle cost of PEV batteries, avoid material shortages, lessen the environmental impact of new material production, and potentially provide low cost active materials for new PEV battery manufacturing.

III. Approach for Overcoming Barriers/Challenges

U.S. DRIVE focuses on two main areas: 1. Advanced Battery Materials Research and 2. Advanced Battery Cell Research. Within each of these broad areas, research is focused on three chemistry classes: a – Beyond Li-ion (BLI); b – Enhanced Li-ion; and c – Next gen Li-ion chemistries. Next gen and BLI have been described above. Enhanced Li-ion are cells made from materials that are commercialized now, (graphite anode/ nickel, manganese, cobalt oxide cathode (NMC)), but that have improvements to enable features such as XFC, cold temperature performance, and enhanced abuse tolerance.

As mentioned above, major advances have been achieved in the short time that researchers and developers have focused on EV batteries. Over the past several years, DOE has tested EV batteries showing deep discharge cycle life improvement to 1,000 cycles and over the same time period cost has decreased from ~$1,000/kWh to just under $250/kWh. All of these improvements have been achieved using the graphite/NMC class of Li-ion cells. Cycle and calendar life of next gen and BLI chemistries are well short of EV goals. Most cells employing a significant amount of silicon provide (at most) 500 deep discharge cycles and less than two years of calendar life; and BLI cells typically do much worse, with cycle lives on the order of 100 cycles or less. In addition, the characteristics of low temperature performance and extreme fast charge are lacking in all chemistries.

An overview of the technologies and their likely ability to meet the challenging DOE/U.S. DRIVE cost goals are shown in Figure 1. Because of the large variation in the state of development for different battery technologies, the VTO energy storage effort also includes multiple activities focused on developing BLI and next gen materials and cell components, and some synthesis and design R&D to address remaining high cost areas within the entire battery supply chain.

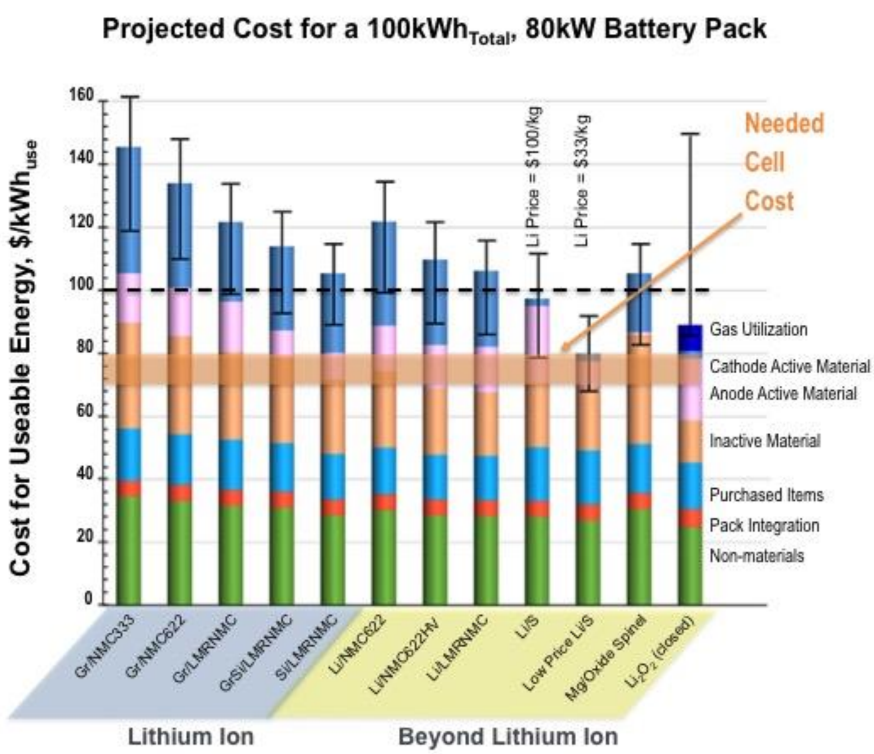


Figure 1. Next Gen Li-ion and BLI Chemistry Cost Estimates with no XFC capability

The potential cost decrease, assuming successful research results, in the three technology areas (BLI, next gen Li-ion, and current Li-ion with no enhancement in performance) over time is shown in Figure 2.

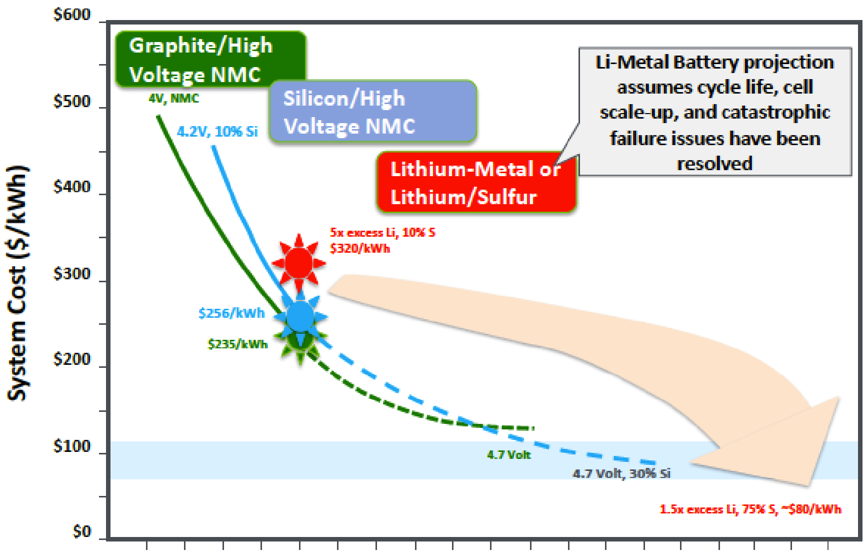
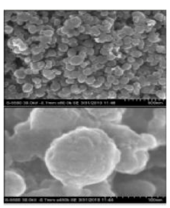


Figure 2. Potential cost trends for Li based EV batteries.

Figure 3 shows an overview of the DOE research focus areas for vehicular energy storage technologies.

Advanced Battery Materials Research
TRL 2-3



Participants

- National Laboratories
- Universities & Industry
- Battery500 Consortium

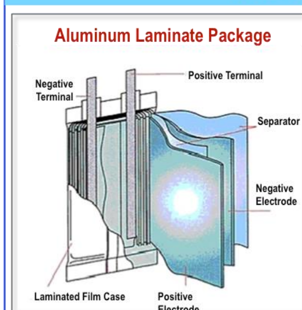
Battery Materials Research Areas

- High capacity/high voltage cathodes
- Alloys & lithium metal anodes
- Material diagnostics and modelling

Battery Materials Targets

- Cathode capacity >300mAh/g
- Anode capacity >1,000mAh/g
- High-voltage cathodes & 5V stable electrolytes
- Solid-polymer electrolytes with $>10^{-3}$ S/cm ionic conductivity

Advanced Battery Cell Research
TRL 3-4



Participants

- USABC
- National Lab Testing
- Industry FOAs
- CAEBAT

Robust EV Battery Cell Development

- Cost reduction
- Power and capacity Improvement
- Cycle and calendar life
- Fast charge and low temperature capability

Battery Pack Targets

- $100/kWh EV pack cost
- 1,000 cycles and 10+ year calendar life
- Fast charge (80% SOC in under 15 min)

Figure 3. Overview of Major DOE Research Areas

In addition to these research areas, DOE also funds supporting R&D that includes testing and requirements and test procedures development. All three are described below.

III.1. Advanced Battery Materials Research

This Advanced Battery Materials Research activity addresses the fundamental issues of chemistries and materials associated with Li batteries. It emphasizes the identification and mitigation of failure modes, coupled with materials synthesis and evaluation, advanced diagnostics, and improved electrochemical models. The current team includes Argonne National Laboratory (ANL), Brookhaven National Laboratory (BNL), Lawrence Berkeley National Laboratory (LBNL), National Renewable Energy Laboratory (NREL), Oak Ridge National Laboratory (ORNL), Pacific Northwest National Laboratory (PNNL), Sandia National Laboratory (SNL), and several universities and commercial entities. Quarterly technical reports for each project in this area are available at http://bmr.lbl.gov/reports.

III.1.a) BLI Materials R&D: While the research on next-gen Li-ion cells is expected to lead to a further decrease in the cost of batteries, further improvement in energy may enable even less expensive batteries. Systems such as lithium-sulfur and lithium metal -high voltage cathode promise significant increases in energy density; however, both of these systems have major issues with both the anode (Li metal) and the cathode (sulfur and high voltage cathode). DOE will fund a number of innovative approaches to enabling these active materials. A sample of those concepts includes:

- Composite solid electrolytes that incorporate a ceramic material to inhibit Li dendrites and a softer polymer to enable Li transport;
- Atomic layer deposition (ALD) treated ceramic electrolytes to reduce the interfacial impedance in solid state batteries;
- Dopants and surface coatings to stabilize high voltage oxide cathodes;
- Electrolyte additives to enable cell operating voltages above 4.5V;
- Polysulfide containment and blocking strategies at the electrode, material, binder, or separator;
- Novel electrolytes that support Li-sulfur cells with more reasonable electrolyte levels, Figure 4; and
- Micron sized structures that isolate the Li metal from excessive interaction with electrolyte, Figure 5.

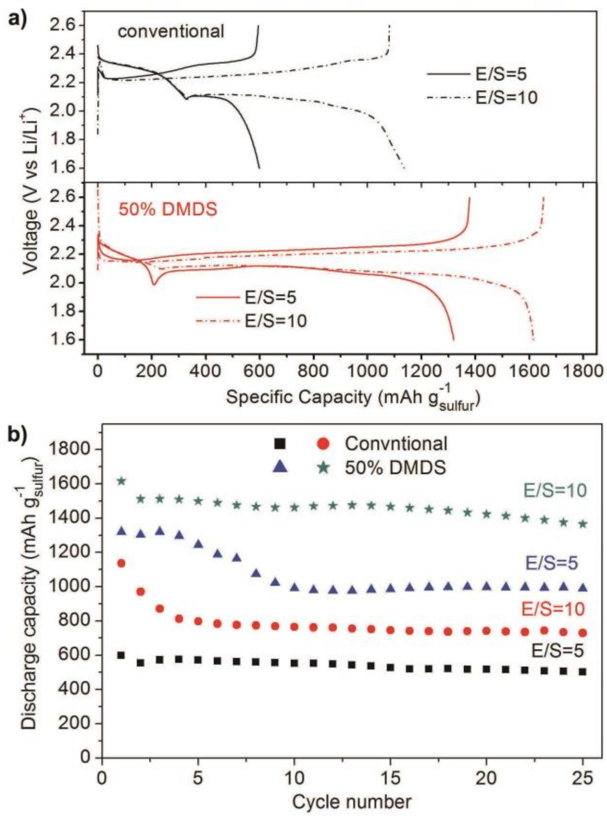


Figure 4. Electrochemical performance of high-sulfur-loading cathode with low electrolyte to sulfur (E/S) ratios of 10 and 5 mL g^{-1} in conventional and DMDS-containing electrolytes. Research from the Wang group at Pennsylvania State University.

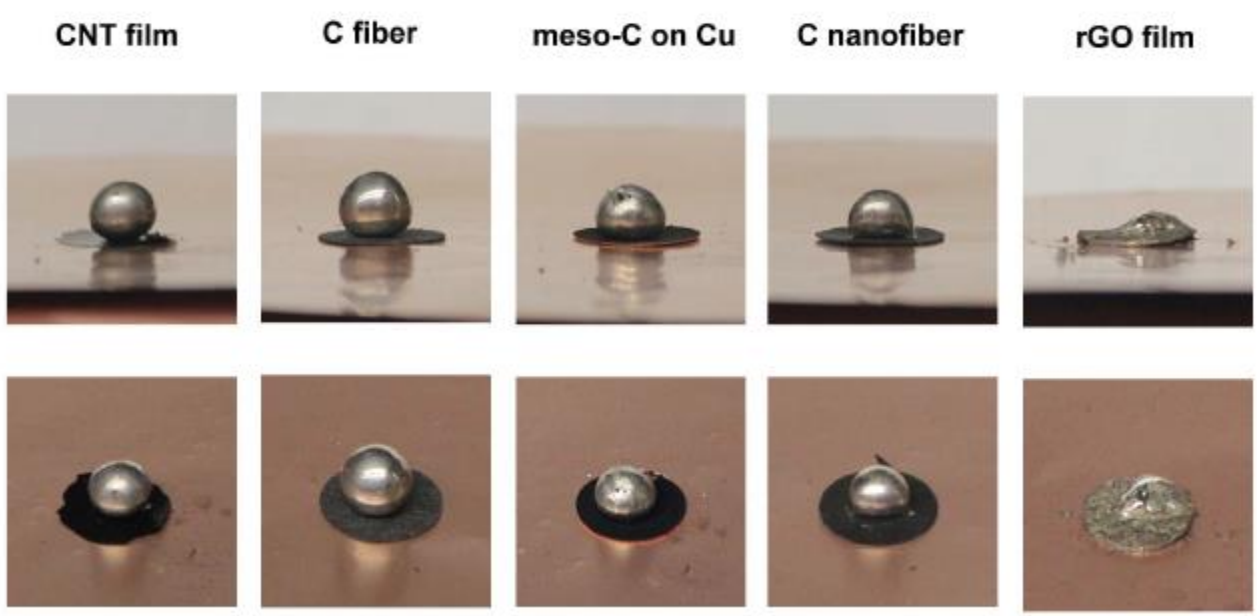


Figure 5. Example of various carbons' ability to incorporate Li metal. r-GO is being used as a Li-metal host. Research from the Cui group at Stanford University.

Battery500 Project: In late 2016, DOE awarded a new project called Battery500, with the goal of developing and demonstrating cells with a specific energy of 500Wh/kg, approximately twice that of today's cells, Figure 6. The project is led by PNNL, with participation from BNL, INL, the SLAC National Accelerator Lab, Binghamton University, Stanford University, University of Texas at Austin, and others. There are two primary technical thrusts being pursued to achieve a 500Wh/kg cell: i) a Li-metal/NMC cell, and ii) a Li-metal/Sulfur cell. Both chemistries can meet the extremely aggressive energy target, with the Li/NMC cell seen as lower energy but lower risk, and the opposite for the Li-sulfur system.

In addition, DOE, in collaboration with the Battery500 team, has recently awarded 15 "seedling projects" that will supplement the work within the Battery500 project. DOE recognizes that no program can identify all of the most promising research avenues, Table 3. Thus, it is hoped that the seedling projects will bring additional ideas for solving the difficult technical issues associated with these cells, and possible new ways of achieving the Battery500 goals.

Strategic Goal	Develop and demonstrate cells with a specific energy of 500Wh/kg and achieving 1,000 cycles
Lead	Pacific Northwest National Laboratory

National Labs	Universities	Advisors
Pacific Northwest National Laboratory	Binghamton University	IBM
Brookhaven National Laboratory	The University of Texas at Austin	Stanford University
INL Idaho National Laboratory	Stanford University	Tesla
SLAC	University of Washington	USABC
	UC San Diego	

Keystone Projects	Lithium metal anode and high nickel NMC cathode	Lithium metal anode and sulfur cathode	Electrode Architecture and Cell Design
Seedlings	New Innovative Research Projects to be added each year		

Figure 6. Overview of the Battery500 Project and its Participants

Table 3. Battery500 Seedling Projects

Organization	Description
Univ. of MD: College Park	Research innovative iron-based materials for high-energy cathodes for high-energy Li-ion battery technologies.
LBNL	Research thick cathodes using freeze casting methods for solid-state lithium batteries.
Penn State University Park	Research multifunctional Li-ion conducting interfacial materials that enable high- performance lithium metal anodes.

Organization	Description
Mercedes-Benz R&D North America, Inc.	Research a scalable synthesis to enable very thin coatings on solid state electrolyte membranes to enable high performance Li-Sulfur Battery.
Univ. of MD: College Park	Using 3D printed, low tortuosity frameworks, develop solid state Li-ion batteries.
General Motors LLC	Design, engineer, develop, and integrate pouch-format cells for lithium-sulfur batteries to achieve high-energy density and long cycle life.
University of Pittsburgh	Research sulfur electrodes utilizing Li-ion conductor (LIC) coatings for high-energy density advanced Li-sulfur (Li-S) batteries.
Cornell University	Research highly loaded sulfur cathodes and conductive carbon coated separators that enable high-energy batteries.
Univ. of MD: College Park	Research advanced electrolytes to limit dendrite growth in Li-metal cells.
Texas A&M Engineering Experiment Station	Utilize analytical and experimental approaches to examine the interface between solid state electrolytes and Li-metal anodes and identify methods for mitigating dendrite growth.
Navitas Advanced Solutions Group, LLC	Research a solvent-free process to fabricate all-solid Li batteries.
Wayne State University	Research novel full-cell, high-energy Li- metal batteries based on 3-dimensional architectures.
Oregon State University	Research and develop a new process to produce Li_2S@graphene composite cathodes to inhibit polysulfides and thus enhance cycle life.
SUNY Univ. at Stony Brook	Research li-sulfur batteries using a novel sulfur rich nanosheet composite cathode.
University of Houston	Research high-energy solid-state Li batteries with organic cathode materials.

III.1.b) Next-gen Li-ion Materials R&D. Investigation will focus on silicon-based alloy anodes, high-capacity and high voltage layered cathodes, and high voltage electrolytes and additives. Over the past five years, a consensus has been reached in both advanced battery research organizations and in industry that the fundamental issue with silicon-based Li-ion batteries is the instability of the silicon solid electrolyte interphase (SEI). The SEI is a film that forms on the anode active particles that inhibits or stops further reactions between the extremely low voltage lithiated anode and the electrolyte. Without this film, or with a film that is not sufficiently passivating (as in silicon), these reactions proceed continuously, consuming Li and leading to rapid capacity fade and short cell life. In graphite based cells, this passivating layers is robust and long-lived, giving rise to batteries that can deliver thousands of cycles and last for 15+ years. This is not the case with silicon containing anodes.

An overview of recent performance metrics of silicon containing cells is shown in Figure 7. As mentioned above, many of the issues impacting silicon-based Li-ion cell life are rooted in the unstable SEI, those include low cycle and calendar life, poor coulombic efficiency, and large first cycle irreversible loss.

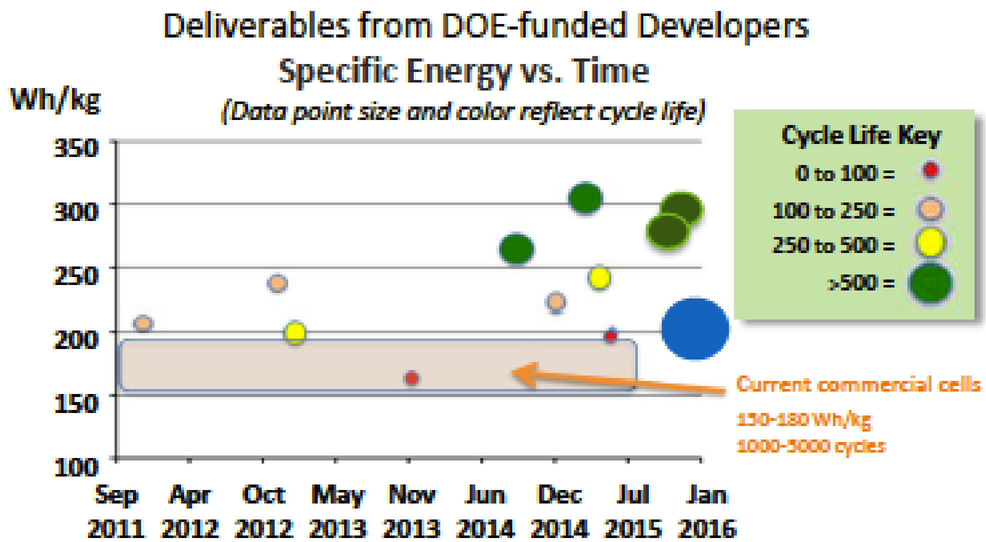


Figure 7. Specific energy and life of silicon containing cells

SEI Stabilization (SEISta) Project: With the realization that the silicon anode SEI is a primary cause of the poor life of silicon-containing next gen Li-ion cells, DOE has formed a project called SEISta, for SEI Stabilization. The aim of this project is to fundamentally understand the passivation film on silicon model anode systems. NREL leads this activity, with support from ANL, LBNL, ORNL, and SNL. The program is developing model silicon anode systems, both with and without surface modifications (including SiOx based). With these model silicon materials, and using standard electrolytes, the team uses advanced diagnostic techniques including NMR, Raman and infrared spectroscopy, TEM and SEM microscopies, and others to characterize and understand the SEI. In addition, first principles modeling efforts will be used to help understand the film's components and properties. The result of these investigations will include, but not be limited to:

- An understanding of the components of the SEI on silicon particles both with and without coatings;
- An understanding of how the film changes over time, both with and without cycling;
- Insight into how "contaminants", like transition metals that can dissolve from the cathode and migrate to the anode, can impact the SEI and its characteristics;
- A means to characterize the passivating nature of the film, or how fast it grows over time; and
- A means to characterize the solubility of the film.

To achieve higher energy, research will focus on using silicon and other alloy anodes to their greatest extent. Currently, most cell developers and researchers limit the usable capacity of silicon to the 600-1000mAh/g range to extend its life. To achieve these higher density anodes, a better fundamental understanding of the fade mechanisms associated with higher Silicon utilization must be achieved.

Next-gen Cathode R&D: In addition to high-energy alloy-based anodes, high-energy cathode materials will continue to be researched. A relatively new set of cathode materials that will continue to be pursued are the disordered lithium excess cathodes. These materials can provide high capacity (250—300mAh/g vs. the 150-200mAh/g of traditional layered oxide). This high capacity is attributed to oxygen activity.

Figure 8 shows one example where the tradition transition metal reaction could only provide about 140mAh/g and the rest, shaded in green, is attributed to oxygen activity.

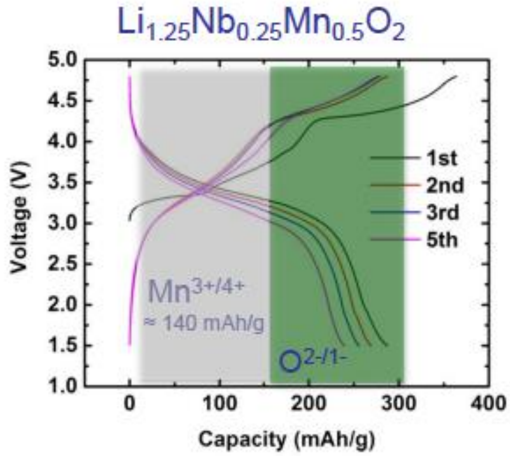


Figure 8. Charge discharge curves a disordered and lithium rich material. Research from the Ceder group at UC Berkeley.

DOE will continue to support efforts to understand and model the life limiting mechanisms in Li based cells. One of the ongoing challenges of verifying a 10 to 15-year life is the need to rely on one to two years of testing data. This effort will continue work toward integrating and improving existing models to understand fade mechanisms and thus develop a more accurate model of battery life. In addition, cost models will be refined and improved to help developers and researchers concentrate on those specific items that will be most effective in reducing battery cost.

III.1.c) Enhanced Li-ion Materials R&D. The baseline system, graphite/NMC, has been successfully commercialized in PEVs and EVs due to its good weight, volume, life, and decreasing cost. However, cold temperature operation and extreme fast charge are still areas needing fundamental improvement. R&D into enabling these capabilities will likely include new active materials' morphologies and sizes, novel electrode designs, new electrolytes with enhanced transport properties, low impedance separators, and possibly enhanced thermal management technologies to manage the heat generated during 400kW charges. XFC cell level requirements are shown in Section III.2.c. Some specific concepts that are being discussed to address these issues include

- Micron-level engineered graphite electrodes where the graphite axis that supports Li diffusion is oriented perpendicular to the current collector and separator to achieve maximum Li conductivity;
- Nano-engineered active particles to ensure that the highest conductivity "face" of each particle is also the largest in area to maximize Li transport;
- Investigation into electrolytes with higher transference number to minimize polarization resistance;
- Numerical and experimental investigation into Li plating and dendrite growth on graphite anodes;
- Electrode additives to enhance electronic conductivity; and
- Investigation into the likelihood of cathode particle cracking during XFC.

For the 12V start/stop system, many developers have proposed lithium titanate (LTO)/lithium manganese oxide (LMO) cells as the most promising. LTO/LMO cells offer excellent power, reasonable energy, and very good abuse. However, issues with low temperature (cold crank) power and both gassing and reduced life at elevated temperatures remain problematic. Some specific concepts that are being discussed to address these issues include:

- Electrolyte additives to enable both improved low temperature power and high temperature stability;
- Surface treatment on LTO particles to mitigate gassing at high temperatures;
- Modified formation procedures to effectively remove species that promote gassing; and
- Doped and coated LMO active particles to mitigate manganese (Mn) dissolution which appears to cause or accelerate LTO gassing at high temperature.

Recycling end of life batteries is a promising route to extremely low-cost battery active material supplies. Several recyclers, developing novel recycling processes under DOE contract, believe they can produce cathode active materials costing 20-50% of pristine materials. The increase in value and cost from raw materials through final electrode construction is significant, a minimum of a factor of two. Research will continue into processes to recover active materials and other valuable cell components in an attempt to produce either refurbished electrodes, or recycled active materials at significantly reduced cost compared to new electrodes or active materials.

Life Cycle Assessments (LCA) will continue to be conducted to understand the overarching impacts of material mining, production, use and battery manufacturing and use on energy, water, and other natural resources. The objectives include: examine material scarcity issues; characterize cradle-to-gate energy and emissions intensity of Li-ion batteries and identify means for their reduction; characterize Li-ion battery recycling; develop improved recycling process to maximize material recovery; determine impact of battery reuse on recycling processes and economics. Sample results from a recent LCA which investigated cradle to grave energy use associated with various battery materials is shown in Table 4.

Table 4. Cradle-to-gate energy intensity (MJ/kg)

Active Material	Cradle-to-Gate Energy Intensity (MJ/kg)
LCO prepared with a solid-state method	180
NCM ($LiNi_{0.4}Co_{0.2}Mn_{0.4}O_2$)	140
LFP prepared hydrothermally ($LiFePO_4$)	60
LMO ($LiMn_2O_4$)	40
Silicon	1000
Lithium	160
Graphite	90

III.2. Advanced Battery Cell Research

This program has the goal of enabling full cell performance using enhanced Li-ion and next gen Li-ion chemistries, and to a lesser extent, BLI, ultimately helping industrial developers of Li-ion batteries overcome key barriers in high-energy EV applications. This involves addressing life, performance, and abuse tolerance issues, and researching and developing promising processing technologies.

III.2.a) Beyond Li-ion Cell Level R&D: Many of the research activities into BLI chemistry will take place at the materials level. But there are also multiple research areas that might be pursued to address cell-level issues. Some of those are:

- Fabrication of high loading sulfur electrodes with good electronic conductivity;
- Sulfur electrode or separator designs to mitigate polysulfide migration issues;
- Design of Li-sulfur cells with minimal electrolyte volume. Near all Li-sulfur cells made today have excess electrolyte to enable dissolution of polysulfides. This excess electrolyte increases cell cost and decreases cell energy; and
- BLI cell and electrode structures that maximize current uniformity to reduce or eliminate dendrite growth.

Finally, there is currently no standard method to detect the growth of Li dendrites inside of a functioning cell. The ability to do this may be critical if cells are to be operated in regions where dendrite growth is possible. Thus, DOE may fund efforts to develop a technique to detect dendrite growth during cell operation. Note that this ability to detect dendrite growth in a functioning cell is critical to use when operating enhanced Li-ion or next gen cells in an extreme fast charge manner, and when monitoring the standard cycling of a BLI cell.

Mathematical simulations will provide novel ways of understanding BLI chemistries. The performance of new materials will be assessed for usefulness in real-world operation by utilizing macroscopic cell-level models. This will allow for an alternative methodology of extrapolating material performance to cell performance without embarking on an extensive battery-manufacturing effort. A specific modeling effort will be undertaken into Li dendrite growth under standard and XFC conditions. The focus of this work will likely be on incorporating real-world conditions, such as electrolyte surface properties, current distributions, and other cell characteristics.

III.2.b) Next Gen Li-ion Cell R&D. Work on next-gen cell issues will focuses on research, development, and engineering of advanced cell chemistries that simultaneously address the life, performance, abuse tolerance, and cost issues associated with cells using alloy-based anodes and/or high voltage cathodes. In recognition of the several issues facing developers of next-gen Li-ion batteries, DOE has formed two large working groups: one to understand and address issues with cells containing high voltage and high capacity cathodes, and a second to evaluate possible technical solutions to silicon-based anode issues, part of the SEISta project described above.

Enabling High-energy-High Voltage Cathodes Project: EV battery performance and cost can be met by cells containing layered-oxide-based cathodes; however, these cells need to be cycled to voltages that exceed 4.5 V vs. Li/Li+. On extended cycling at these voltages, however, capacity loss, impedance rise and voltage fade reduces the cell's energy and power output. The objective of the high-energy high voltage cathode team is to mitigate this performance degradation, thereby reducing the lifetime cost of these high-energy batteries. This team includes representatives from ANL, BNL, LBNL, NREL, ORNL, and the Jet Propulsion Lab. Some approaches to stabilize these materials and extend their life include the use of fluorinated electrolytes and additives, coatings, and concentration gradient structures, and others. Figure 9 shows an example of increased cycle life resulting from the use of a fluorinated electrolyte developed at ANL.

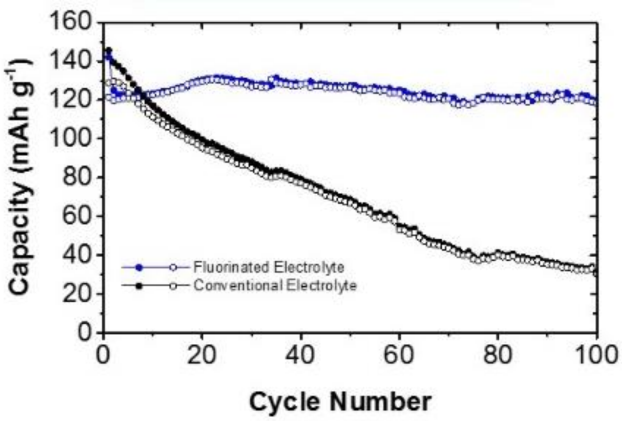


Figure 9. Capacity retention of $LiNi_{0.5}Mn_{1.5}O_4$ /graphite cells cycled at 55 °C, from 3.5-4.9V, with F-electrolyte compared with conventional electrolyte at C/3 rates. (Research from the Zhang group at ANL).

Abuse Tolerance Improvements: SNL leads this task to elucidate degradation mechanisms in enhanced Li-ion, next-gen Li-ion, and potentially BLI cells that lead to poor abuse tolerance (runaway thermodynamics, gas evolution, electrolyte combustion). The response of next gen Li-ion and BLI cells to abusive conditions is largely untested. BLI cells suffer from dendrite growth that can result in internal short circuit and possibly thermal runaway. Silicon-containing cells, in the limited abuse testing conducted thus far, do not appear to be significantly more reactive than traditional Li-ion cells, but may generate more gas.

As developers and researchers push oxide cathodes to higher and higher voltages to improve cell energy, the reactivity of those cathodes' surface species with electrolyte increases, as does the loss of oxygen from the cathode. That lost surface oxygen can then react with the electrolyte releasing large amounts of heat. Techniques being pursued to reduce or eliminate these reactions include cathode surface coatings, concentration gradient cathodes, less reactive electrolyte species, and other techniques. The increased reactivity of the cathode surface with electrolyte is also a major contributor to rapid cell capacity fade, so the techniques used to address the abuse tolerance issue may also result in improved life.

SNL will supplement their abuse testing role by developing and evaluating advanced materials (or materials combinations) that will lead to more abuse-tolerant next gen Li-ion and BLI cells. Abuse modeling at other national labs could provide insight on the major causes of the abuse behavior and how they could be prevented with materials and cell designs.

III.2.c) Enhanced Li-ion Cell R&D

Extreme Fast Charge: It is recognized that a combination of fast chargeable batteries and a network of high capacity chargers can minimize range anxiety and promote the market penetration of EVs and increase total electric miles driven. DOE, in collaboration with the Unite States Advanced Battery Consortium (USABC), drafted requirements for EV cells having XFC capabilities,

Table 5. Compared to the non-XFC EV requirements, the XFC requirements permit a lower energy density and specific energy along with a slightly reduced calendar life (from 15 to 10 years) to encourage fast charge capability while maintaining the same aggressive cost target of $75/kWh.

Table 5. Subset of performance and cost targets for XFC capable EV cells. Table cells are color coded to indicate where current batteries fall short of targets.

End of Life Characteristics at 30°C	Cell Level
Cost	\$75/kWh
Fast High Rate Charge	80% ΔSOC in 15 min
Peak Specific Discharge Power, 30 s Pulse	700 W/kg
Peak Specific Regen Power, 10 s Pulse	300 W/kg
Useable Specific Energy @ C/3	275 Wh/kg
Useable Energy Density @ C/3	550 Wh/l
Calendar Life	10 Years
DST Discharge Throughput for a 50kWh Battery	50 MWh

In early 2017, DOE assessed the knowledge base of the fast charging capability of automotive batteries; used that to identify technical gaps for fast charging; and identified R&D needs. The results of this study are being published in a report titled "*XFC Technology Gap Assessment*" which addresses issues with batteries, power electronics, the grid, and all other aspects of enabling XFC in electric vehicles. The main battery issues identified regarding fast charging were:

- Higher cost and lower energy density cells: 100% more expensive and with lower energy density compared to today's Li-ion cells that don't support fast charge (due to the need for much thinner electrodes to avoid Li plating), Table 6;
- Cycle life and durability of cells;
- Li plating or deposition (Figure 10) that occurs on the anode above a threshold current density which is approximately 1C for a 4mAh/cm^2 electrode (or 4mA/cm^2); and
- Cell temperature rise during charge.

Table 6. Cell characteristics as a function of charge time

Charging Time, ΔSOC=80%, min	***8***	***10***	***23***	***47***	***53***	***61***
Charger Power Needed, kW	601	461	199	100	88	77
Anode Thickness, µm	**14**	**19**	**43**	**87**	**98**	**103**
Cell cost relative to cell providing 61 min charge	**2.22**	**1.90**	**1.28**	**1.04**	**1.01**	**1.00**

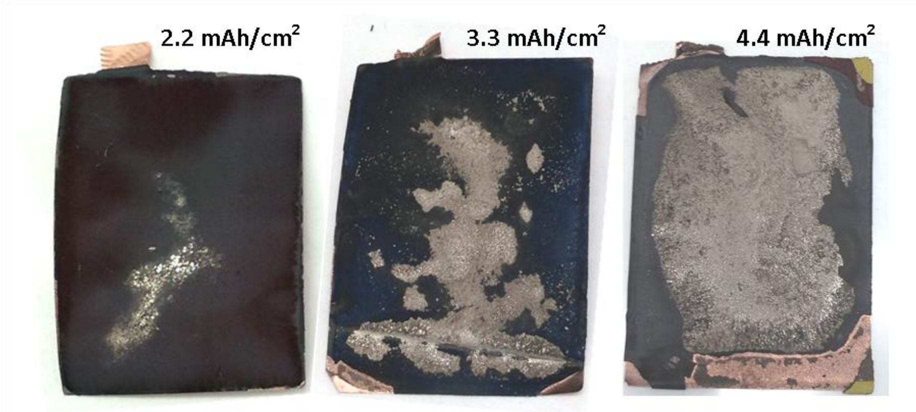


Figure 10. Images of graphite electrode in NMC622/Gr pouch cells. Lithium plating appears as metallic deposits on the surface of the electrode. Research from the Bloom group at ANL.

To address these issues, DOE will investigate a number of approaches, including nano-engineered active particles to enhance both ionic and electronic conductivity, novel electrode structures, electrolytes with higher transference number and conductivity, novel cell, module, and pack designs with enhanced thermal conductivity, and techniques to lessen the likelihood of dendrite formation during fast charge.

Low Temperature Performance: It is widely recognized that Li-ion batteries lose both energy and power when operated at low temperature, <10°C. There are reports of Nissan Leaf EVs traveling only half of their rated mileage when the temperature drops below 0°C. Many of the approaches that are expected to enhance extreme fast charge are also likely to help low temperature performance. Another area that will be investigated will be electrolytes that provide a wider operating range of temperatures than those currently employed.

Lower Cost Material Synthesis and Electrode Design R&D: In 2013 and 2015, DOE funded several electrode design and synthesis R&D projects. These were initiated following the realization that major advances were being made in the lab and by industry in extending the life of Li-ion cells and in formulating full systems that provided 200-300 mile range; however, the cost of making the active materials, electrodes, and other components remained high. Thus, a primary objective of these projects was to develop and demonstrate faster and lower cost synthesis methods. A secondary objective was to enable new electrodes or materials from next-gen materials like silicon.

Research will also be conducted into potentially new cell, module, and system designs that minimize the creation of hazardous waste, the use of energy during their manufacturing processes, and that are most amenable to recycling or reuse.

Several additional research topics that may be pursued include:

- *Particle size and shape control.* This research will concentrate on quantifying the changes in performance with particle size and shape, and on developing processes for manufacturing the materials with the desired size and shape. This is one of the research areas that could improve the performance of existing materials to enhance their ability to be operated at low temperature or at extremely high charge rates.

- *Thin film Li metal production.* One of the advantages of Li metal anodes is their high capacity per unit weight and volume. In practical cells, the anode and cathode must be balanced or nearly balanced. With BLI cathode materials expected to provide areal capacities of 8mAh/cm^2, and assuming a 1.5/1 n/p ratio, one desires Li metal films of 60 microns or less. Current methods of producing such thin Li metal foil are expensive, and it is difficult to handle such thin sheets of the extremely pliable metal. Thus, novel processing and handling procedures may be researched.

- *Three-dimensional electrode designs.* There are many issues with high-energy cell design that could be addressed through the use of 3D or non-planar electrode construction. Electron transport, fast ion transport, higher loading electrodes, are all parameters that could be improved through the use of 3D or non-planar electrode designs.

- *Novel system packaging.* Investigate new system designs that provide enhanced thermal management, reduced cost, weight, and volume. Approaches that eliminate or significantly reduce the need for cell packaging will be explored, especially in the 12V system designs. Novel cooling liquids that may be needed for XFC functionality and for under-hood 12V systems, and possibly low-cost manufacturing technologies to reduce the cost of those liquids, will be investigated.

In addition, DOE works on advanced cell R&D with industry through the USABC and through direct contracts with battery and material developers. The USABC provides technical and programmatic management of the projects under that organization, for which the cost is shared by the developers at a minimum level of 50 percent.

III.3. Requirements and Test Procedure Development and Execution

All of the research and development described above is geared towards meeting and exceeding performance and cost goals. The activities begin by establishing, with close coordination between industry, DOE, and national laboratories, technical requirements for the energy storage technologies and then by developing test procedures that measure progress, in an independent and quantitative manner, against those requirements. When researchers or battery developers produce cells, the national labs conduct standardized tests to enable apples to apples comparisons across organizations and technologies. The requirements and test procedures can be found on the USABC website at http://www.uscar.org/guest/teams/12/U-S-Advanced-Battery-Consortium-LLC.

In the near term, U.S. DRIVE will update the fast charge requirements and develop new test procedures to evaluate existing and emerging technologies and their ability to meet those requirements. In addition, the low temperature performance requirements for EVs, and the associated test procedures, will be reviewed and, if necessary, updated. The main organizations that will spearhead this work are: ANL, INL, NREL, the DOE extreme fast charge project lead, and all U.S. DRIVE members.

IV. Conclusion

The DOE/U.S. DRIVE energy storage R&D effort is focused on overcoming the remaining technical and cost barriers to commercialization of advanced low-cost batteries for EVs and other applications. Table 7 shows an overview of the R&D efforts to be pursued in order to reach the aggressive U.S. DRIVE targets.

Table 7. Overview of energy storage R&D plans

Beyond Li-ion	Next gen Li-ion	Enhanced Li-ion
1: Advanced Battery Materials Research		
Li dendrite suppressing electrolyte additives, polysulfide trapping and binding agents. Novel solid and mixed ceramic/polymer electrolytes. Improve Li dendrite modeling.	Next gen anodes (silicon based) and cathodes, high voltage and non-flammable electrolyte development.	Nano-and micron level engineered active particles, new conducting additives, new electrolytes with high transport numbers,
Li-Sulfur, Li-air materials development; thin film Li production methods; novel electrode designs and particle shapes for polysulfide management. Thin film Li metal processing methods.	High voltage and disordered Li rich oxides, non-flammable electrolytes, high loading electrodes. Particle size and shape control R&D. High performance computing for improved battery materials.	Higher porosity separators for enhanced rate, electrolytes with wider temperature operating window for low T performance, New cell architecture for enhanced rate and thermal performance for 12V and PEV batteries.
Li-Sulfur, Li-air materials development; fast charge and discharge BLI cell and material designs. Thin film Li metal processing methods.	Higher voltage and disordered Li rich oxides, higher Ni content NMC cathode materials, higher voltage and non-flammable electrolytes, high loading electrodes, novel cell designs.	Lighter and thinner cell support materials (such as separator and current collectors) for enhanced rate and low T performance. Three-dimensional electrode designs.
2: Advanced Battery Cell Research		
Develop Li metal dendrite mitigation approaches include electrolyte additives, Li hosting structures, ceramic and mixed ceramic electrolytes, Li metal alloys. Sulfur electrode designs to mitigate polysulfide issues and poor rate. Separator & anode designs to mitigate polysulfide issues.	Understand alloy (silicon)-anode solid electrolyte interphase (SEI), its stability in baseline electrolytes, and the impact of electrolyte additives. High voltage layered oxides or disordered oxide cathodes providing 250mAh/g. Electrolytes with better compatibility with alloy anodes and high V cathodes.	Understand limits to XFC and cold temperature performance targets, 12V cold crank with high temperature life. First gen low temperature enhancements and dendrite mitigation techniques are tested. Novel cell and module designs for 12V systems.
Develop commercially viable BLI cell designs. Current designs often utilize excessive electrolyte (in the case of Li-sulfur cells) or excessive Li metal (in all BLI cells). Evaluate novel Li film manufacturing techniques to reduce the cost of very thin Li metal foils.	Develop alloy anode-based electrode designs to enhance SEI stability. Volume change mitigation technologies. Test XFC and cold temperature performance enhancement technologies on cells. Understand abuse tolerance characteristics of silicon-anode/high voltage cathode cells.	Build cells using novel electrode and cell designs to enhance thermal management, electrical and ionic conductivity, for XFC capability. Investigate abuse tolerance improvement through cell and electrode designs.
Li-Sulfur, Li-air cell integration and optimization; new cell and battery architectures.	Develop novel cell or battery designs that meet life requirements. Integrate novel electrode and active material designs into anode and cathode. Address remaining abuse tolerance issues at the cell level.	Integrate promising electrode & material advances to achieve XFC and cold temperature performance with reduced system cost. Improve abuse tolerance of enhanced Li-ion cell and electrodes.