USDRIVE

DRIVING RESEARCH AND INNOVATION FOR
VEHICLE EFFICIENCY AND ENERGY SUSTAINABILITY

Electrochemical Energy Storage Technical Team Roadmap

June 2013

This roadmap is a document of the U.S. DRIVE Partnership. U.S. DRIVE (Driving Research and Innovation for Vehicle efficiency and Energy sustainability) is a voluntary, non-binding, and nonlegal partnership among the U.S. Department of Energy; USCAR, representing Chrysler Group LLC, Ford Motor Company, and General Motors; Tesla Motors; five energy companies — BP America, Chevron Corporation, Phillips 66 Company, ExxonMobil Corporation, and Shell Oil Products US; two utilities — Southern California Edison and DTE Energy; and the Electric Power Research Institute (EPRI).

The Electrochemical Energy Storage Technical Team is one of 12 U.S. DRIVE technical teams ("tech teams") whose mission is to accelerate the development of pre-competitive and innovative technologies to enable a full range of efficient and clean advanced light-duty vehicles, as well as related energy infrastructure.

In March 2012, DOE announced a 10-year vision for plug-in electric vehicles (PEVs), called the "EV Everywhere Grand Challenge." EV Everywhere aims to enable American innovators to rapidly develop and commercialize the next generation of technologies to achieve the cost, range, and charging infrastructure necessary for widespread PEV deployment. As demonstrated in its guiding Blueprint document, EV Everywhere aligns with U.S. DRIVE technical areas focused on electrochemical energy storage, electrical and electronics, materials, vehicle systems and analysis, and grid interaction (for more information, please see www.vehicles.energy.gov/electric_vehicles/10_year_goal.html).

For more information about U.S. DRIVE, please see the U.S. DRIVE Partnership Plan, www.vehicles.energy.gov/about/partnerships/usdrive.html or www.uscar.org.

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Team Mission and Scope

This U.S. DRIVE electrochemical energy storage roadmap describes ongoing and planned efforts to develop electrochemical energy storage technologies for plug-in electric vehicles (PEVs). The Energy Storage activity comprises a number of research areas (including advanced materials research, cell level research, battery development, and enabling R&D which includes analysis, testing and other activities) for advanced energy storage technologies (batteries and ultra-capacitors).

Previous work on hybrid electric vehicle (HEV) batteries has yielded a relatively mature generation of vehicles (over 400,000 HEVs were sold in the United States in 2012). Over the past five years, the focus of battery development has shifted to higher energy systems specifically suited for Plug-In Hybrid Vehicles and Electric Vehicles (note: Plug-In Hybrid Vehicles or "PHEVs" and All Electric Vehicles or EVs will be referred to as PEV in this document). Significant barriers remain for meaningful market penetration of PEVs. First and foremost, the cost of higher energy batteries remains prohibitive and hampers PEVs from competing effectively with conventional vehicles. Longer term R&D will focus on research in the area of extremely high energy battery chemistries for use in PEVs, and will also support the development of transformational technologies that can significantly reduce the cost of high power systems for HEVs.

The focus of U.S. DRIVE battery R&D activity is to develop the technologies that will reduce battery costs from their current $500-600/kWh to $125/kWh. In addition, vehicle design optimization and performance is often hindered by the size and weight of the battery. Current battery technology is very far from its theoretical energy density limit. In the near-term (2012-2017), with advances in lithium-ion technology, there is an opportunity to more than double the battery pack energy density from 100 Wh/kg to 250 Wh/kg through the use of new high-capacity cathodes, higher voltage electrolytes, and the use of high capacity silicon or tin-based intermetallic alloys to replace graphite anodes. Despite current advances, much more R&D will be needed to achieve the performance and lifetime requirements for deployment of these advanced technologies in PEVs. In the longer term (2017-2027), "beyond Li-ion" battery chemistries, such as lithium-sulfur, magnesium-ion, zinc-air, and lithium-air, offer the possibility of energy densities that are significantly greater than current lithium-ion batteries as well as the potential for greatly reducing battery cost. However, major shortcomings in cycle life, power density, energy efficiency, abuse tolerance, and manufacturing cost currently stand in the way of commercial introduction of state-of-the-art "beyond Li-ion" battery systems. Breakthrough innovation will be required for these new battery technologies to enter the PEV market.

U.S. DRIVE Partnership Energy Storage Goals and Performance Targets

The broad needs of the automotive industry range from relatively small 12 V systems intended for "Start/Stop" applications, through moderate energy and high power HEV batteries, to the larger energy and power batteries needed for PEVs. This diverse set of needs will not be met by a single battery chemistry or design. Each application requires strategic tuning of performance metrics through focused design changes. A set of performance targets has been set in order to reach the goals for start/stop, HEV, and PEV battery systems. A subset of these targets is shown in Tables 1 and 2.

Table 1. U.S. DRIVE End of Life Targets for PHEV Systems and Cells

Battery System Level Goals	PHEV 40
Characteristic	
Available energy (kWh)	11.3
System weight (kg)	120
System volume (l)	80
Discharge power (kW, 10 sec)	50
Regen. power (kW, 10 sec)	25
Recharge rate (kW)	1.4-2.8
Cold cranking power @ -30C (2 secs) (kW)	7
Calendar life (years)	15
Cycle life (cycles)	5,000 deep discharge
Operating temp. range (°C)	-30 to +52
Selling price @ 100k units/year, $	3,400

Cell Level Goals	PHEV40
Characteristic	
Specific energy (Wh/kg)	200
Energy density (Wh/l)	400
Specific discharge pulse power (W/kg)	800
Discharge pulse power density (W/l)	1,600
Recharge time (hours)	3-6
Specific regen. pulse power (W/kg)	430
Regen. pulse power density (W/l)	860
Calendar life (years)	15
Cycle life (cycles)	5,000
Operating temp. range (°C)	-30 to +52

U.S. DRIVE has also recently established system and cell level EV goals, shown in Table 2.

Table 2. U.S. DRIVE EV System and Cell Level End of Life Goals

Energy Storage Goals	EV Battery	EV Cell
Characteristic		
Available energy (kWh)	45	NA
Discharge power density (W/l)	1,000	1,500
Specific discharge power (W/kg)	470	700
Specific regen. power at 20% DOD, 10 sec (W/kg)	200	300
Energy density @ C/3 discharge rate (Wh/l)	500	750
Specific energy @ C/3 discharge rate (Wh/kg)	235	350

Table 2. (Cont.)

Energy Storage Goals	EV Battery	EV Cell
Calendar life (years)	15	15
Cycle life to 80% DOD (cycles)	1,000, deep discharge	1,000, deep discharge
Operating temperature range (°C)	-30 to +52	-30 to +52
Selling price @ 100k units/year, \$/kWh	125	100
Recharge time (hours)	<7, J1772	<7, J1772
Fast recharge time	80% ΔSOC in 15 minutes	80% ΔSOC in 15 minutes

The energy storage targets have been developed through a consensus of the U.S. DRIVE Electrochemical Energy Storage Tech Team (EESTT) consisting of government and industry participants. See http://www.uscar.org/guest/article_view.php?articles_id=85 for more energy storage goals.

Key Issues and Challenges

The U.S. DRIVE R&D portfolio is focused on overcoming a number of fundamental technical problems to achieve the aggressive but achievable goals shown above. The major challenges to developing and commercializing batteries for PEVs are as follows:

Cost

The current cost of high-energy Li-ion batteries is approximately \$500-\$800/kWh, a factor of four to six times too high on a kWh basis. The main cost drivers are the high cost of raw materials and materials processing, the cost of cell and module packaging, and manufacturing costs. Cost is not the sole barrier; however, it is an overriding factor for market success in conjunction with other technical targets (e.g., life, weight, and volume). Addressing the cost barrier requires developing and evaluating lower-cost components including much higher energy active materials, alternate packaging, and processing methods; and joint work with U.S. suppliers to implement these low-cost solutions.

Performance

Higher energy densities are needed to meet both volume and weight targets for PHEV and EV applications. Current batteries are approximately a factor of two to three times too heavy and large compared to the 40-mile PHEV requirements. An increase in energy density will also reduce the amount of material and supporting hardware needed to construct the entire battery and will thus reduce battery costs. An additional barrier to EV battery commercialization is low temperature performance, which is particularly critical when the battery is the sole power source.

Abuse Tolerance, Reliability and Ruggedness

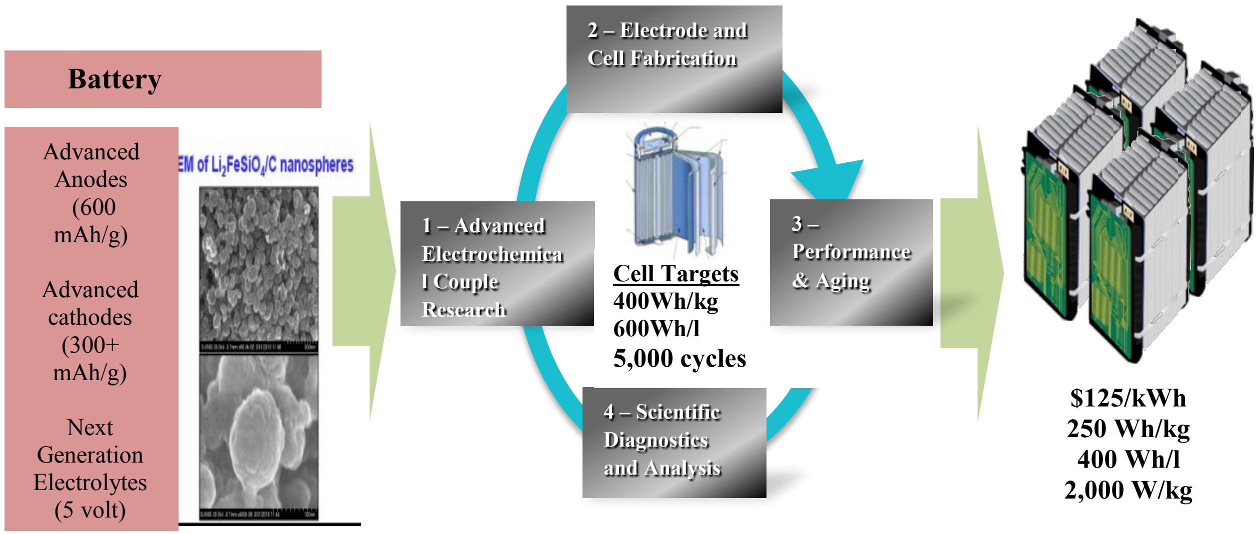
It is critical that any new technology introduced into a vehicle be abuse tolerant under both routine and extreme operating conditions. Many lithium batteries are not intrinsically tolerant to certain abusive conditions. The use of large format lithium cells increases the urgency with which these issues must be addressed. In addition, current thermal control technologies, although adequate to dissipate heat in today's systems, are too expensive and significantly add to the systems' weight and volume.

Life

The barriers related to battery life are the loss of available power and energy due to use and aging, and the challenges of accurate life prediction. For high-energy batteries in a PEV application, a combination of energy and power fade over life are challenging issues as the battery must provide significant energy over the life of the vehicle and either provide full vehicle power (for an EV) or high-power HEV pulses (for a PHEV) near the bottom of its State of Charge (SOC) window. Today, batteries designed for HEVs can deliver 300,000 shallow discharges. However, batteries with a higher energy density have difficulty meeting the 5,000 deep discharge cycle requirement for PHEVs.

Strategy for Overcoming Barriers/Challenges

The scope of U.S. DRIVE energy storage R&D includes focused fundamental materials research, generally spearheaded by the national laboratories and universities, and battery cell and pack development and testing, mainly by commercial developers and national laboratories. Figure 1 illustrates one of the general approaches being used to develop higher energy battery materials, improve battery power, durability, and abuse tolerance, and to significantly reduce cost. In addition to battery materials and design improvements, the Partnership is focused on less expensive manufacturing techniques, advanced thermal management technologies, novel packaging, and computer aided engineering battery design tools.



Battery Materials Research addresses fundamental issues of materials and electrochemical interactions associated with advanced batteries. It attempts to develop new materials, to use advanced models to predict failure modes, and scientific diagnostic tools to gain insight into why materials fail and to develop ways to mitigate that failure.

Advanced battery research focuses on the optimization of new cell technologies that incorporate new battery materials. The emphasis is to identify, diagnose, and mitigate issues that impact the performance (power and energy) and life of cells that contain new cathodes coupled with advanced electrolytes and anodes.

Advanced Battery Development R&D focuses on the development of robust cells and modules to significantly reduce battery cost, increase life, and improve performance. This research aims to ensure these systems meet specific goals for particular vehicle applications.

Figure 1. The U.S. DRIVE Partnership Battery R&D Approach and Process

Research and Development Roadmap

PEVs introduced in recent years (like the Chevy Volt, Ford CMAX, and Nissan Leaf) are powered by first generation lithium-ion traction batteries. Typically, these feature a graphitic anode coupled with a cathode of layered oxides or spinels, and a standard lithium electrolyte system ($LiPF_6$ salt and a blend of carbonates with various additives). Typical first generation Li-ion cells have a nominal energy of 150Wh/kg and 250Wh/l at the cell level; battery system level performance is about 50% lower.

The heightened focus on the larger and more powerful PEV batteries requires a second generation of Li-ion technology. Since these batteries carry significantly more energy, they are more heavily penalized for cost, weight, and volume, so it is necessary to double power and energy and decrease cost by 70% to be achieve a marketable product. Over the past five years, DOE has tested batteries showing deep discharge cycle life improvement from 1,000 cycles to over 3,500 cycles. In addition, over the same time period, cost has decreased from over $1,000/kWh to just under $600/kWh. A few major battery manufacturers have predicted a further cost decrease to the $400/kWh range in the near future. However, further improvements are needed.

Higher energy and higher power electrode materials promise to significantly lower battery cost by reducing the amount of material and the number of cells needed for the entire battery pack. Work is needed to develop new materials and electrode couples that offer a significant improvement in either energy or power over today's technologies. Some specific technologies of interest include, but are not limited to: the design and development of 2nd generation lithium ion batteries that contain high voltage (5V) and/or high capacity (>300 mAh/g) cathode materials; the design and development of 3rd generation lithium ion batteries that contain advanced metal alloy and composite anodes such as silicon carbon that offer two to four times the capacity of today's graphite anodes; and high voltage and solid polymer composite electrolytes. Also, efforts must include the development of novel electrolyte formulations and additives to form a stable solid electrolyte interphase for improved abuse tolerance, longer life, low temperature operation, and fast charge capability. Going forward, a larger portion of battery research will be on beyond lithium ion battery technologies that offer the potential for very high-energy and low cost such as solid-state (lithium metal with solid electrolytes), lithium sulfur and lithium air batteries. All of these promise theoretical energy densities from two to five times that of traditional lithium ion. In addition, some non-lithium couples (e.g., magnesium, zinc) may show promise in the long-term. Research is needed to advance these next generation technologies from the university and national laboratory arena to the first stages of industry development through the development and testing of full cells. Table 3 shows the relative attributes of the technologies being investigated.

Table 3. Demonstrated Attributes of Representative Battery Technologies

		Battery Performance (Pack Level)				
	Maturity	**Specific Energy (Wh/kg)**	**Energy Density (Wh/l)**	**Power (W/kg)**	**Current Life (cycles)**	**Abuse Tolerance**
Lithium-ion (current status)	Pack	50-80	100-150	500-750	>5,000	Meets SAE J2929
Lithium-ion (future generations)	Cell 20Ah+	**155**	**205**	**800**	**~500+**	TBD
Lithium metal polymer (solid)	Cell 20Ah+	150	250	**<100**	**~1,000**	TBD

Table 3. (Cont.)

		Battery Performance (Pack Level)				
	Maturity	**Specific Energy (Wh/kg)**	**Energy Density (Wh/l)**	**Power (W/kg)**	**Current Life (cycles)**	**Abuse Tolerance**
Lithium metal/sulfur	Cell (Lab)	**250-400**	**180-250**	**<100**	**~100**	**Concern**
Lithium metal/air	Lab Devices	400-600(?)	200(?)	**Poor**	**?**	**Concern**

High-power battery development may be supported on transformational technologies with the potential to significantly reduce the cost of HEV and microhybrid vehicle batteries. The work will focus on the development of robust prototype cells that contain new materials and electrodes that offer a significant reduction in battery cost over existing technologies. Research will be conducted to expedite the development of more efficient electrode and cell designs and fabrication processes to reduce the cost of high-volume production of large format lithium-ion batteries. Pack-level innovations will focus on technology to reduce the weight and the cost of thermal management systems, structural and safety components, and system electronics. Currently, these “non-active” components of a battery increase the volume, weight (approximately 70 percent by weight of the battery), and cost of the finished product. Approaches to reduce the size of these inactive components in the cell and battery will be pursued.

U.S. DRIVE will also accelerate the market entry of advanced batteries by supporting the scale-up, pilot production, and commercial validation of new battery materials and processes. New materials for advanced cathodes, anodes, and electrolytes are being developed by universities, national laboratories, and industry, but the commercial scale-up of such materials is often limited in scope. Studies of recycling and reuse of lithium batteries will continue. This activity will also continue to validate requirements and refine standardized test procedures to evaluate battery performance and life, as well as identify areas requiring additional R&D.

The remainder of this document is organized into two areas: Next Generation Li-ion Materials and Batteries, and Beyond Li-ion Materials and Batteries. Next Generation Li-ion refers to batteries that DO NOT contain a Li metal anode, but which instead include graphite-based, graphene-based, Si-based, and other non-Li metal anode materials. Beyond Li-ion refers to Li metal-based batteries such as Li-sulfur, Li-air, and non-Li containing batteries such as Na, Zn, or Mg-ion based devices.

Next Generation Lithium-Ion Battery R&D

This area’s goal is to advance the performance of materials, designs, and processes that significantly improve the performance and reduce the cost of Li-ion batteries using a non-metallic anode. Specific areas of investigation include high-energy anodes (e.g., containing Si or Sn), high voltage cathodes, high voltage and non-flammable electrolytes, novel processing technologies, high energy and low cost electrode designs, and others. This work spans the entire range of U.S. DRIVE activities, from advanced materials R&D to battery development. Samples of that work are provided below; more complete descriptions are available in the VTP annual progress reports available at http://www1.eere.energy.gov/vehiclesandfuels/resources/fcvt_reports.html.

High Voltage Cathodes

The work on advanced cathodes is primarily focused on the Li-Mn rich oxide materials of general formula $x\mathrm{Li_2MnO_3}\bullet(1\text{-}x)\mathrm{LiMO_2}$ (M = Ni, Mn, Co), the 5V spinel materials ($\mathrm{LiMn_{1.5}Ni_{0.5}O_4}$), traditional NMC operated at higher voltages, and, to a lesser extent, on the higher voltage silicates and phosphates. Figure 2 shows the theoretical specific energies of some of the main cathode materials under investigation.

Recently, a team was organized to evaluate a graphite/high voltage electrolyte/$\mathrm{LiMn_{1.5}Ni_{0.5}O_4}$ battery. The use of a high-voltage Ni-Mn spinel is hindered by the oxidative instability of the electrolyte, Mn dissolution, and poor rate performance over time. Researchers studied surface coatings, electrolyte purity, additives in solution to make better SEIs, and additive/binder free operation. This rounded approach helped researchers to not only assess the ability of this chemistry to meet the PHEV targets, but also allow new methods and technologies to be developed. Other projects in the program include studies of spray pyrolysis as a cathode material production method; doping of traditional NMC materials; stability of inactive components (like current collector, binder, and conductive additive) at high voltages; computational searches for new cathode materials; in-situ investigations into solvothermal synthesis reactions; and Li bearing mixed polyanion glasses.

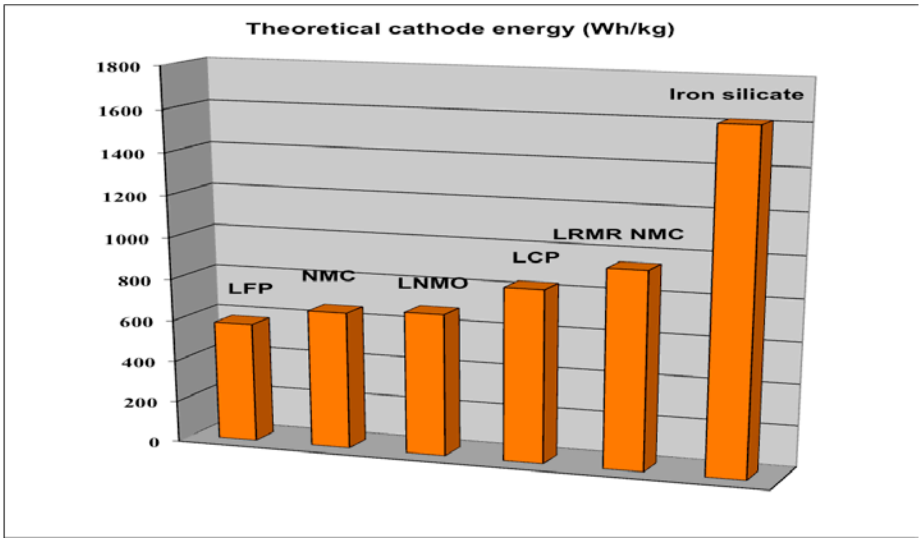


Figure 2. Theoretical Cathode Energy Densities (LFP = Li iron phosphate, NMC = nickel, manganese, cobalt oxide, LNMO = 5V Ni Mn spinel, LCP = lithium cobalt phosphate, Li-Mn rich oxides)

New High Voltage/High Capacity Cathodes

Continued research and development is planned on Li-Mn rich oxide materials where focus will remain on issues such as voltage fade, high impedance especially at low state of charge, metal dissolution leading to anode impedance rise, and low electrode density leading to low $\mathrm{mAh/cm^2}$. Work will also continue on extending the voltage range of the traditional NMC cathode. As the voltage is increased in these materials, one achieves both increased capacity (mAh/g) and increased energy via the increased voltage. Approaches to enabling this higher voltage operation include varying the material composition from the inside to the outside of the particle (with the outer material being more stable against the electrolyte), coatings, doping, and electrolyte additives that form a protective coating on the cathode particles.

Other more exploratory materials to be pursued include high voltage phosphates, such as Li manganese phosphate ($\mathrm{LiMnPO_4}$) and cobalt phosphates ($\mathrm{LiCoPO_4}$). These materials typically have poor power performance and also may require stabilization of their surfaces against the electrolyte. Iron and manganese silicates (e.g., Li_2FeSiO_4) offer extraordinarily high energies (5V and 330 mAh/g), more than double that of NMC. However, to date researchers have not succeeded in reversibly extracting the second Li from these materials, and high voltage operation will also be a challenge.

High Energy Alloy Anodes

In June 2010, a focus group was organized to develop a Si-based anode with low 1st-cycle loss (ICL<15%) and high cycling efficiency (>99.99%) operating at 1,200 mAh/g. The projects currently underway include: Cu foam current collectors to enable better utilization of Si nanoparticles; Si nanowires directly deposited on current collectors; a variety of nano-structured and nano-porous Si materials; a Si/graphene composite; a SiO_x material; a Si-PAN hybrid material; Si clathrate materials; a family of MXene materials such as Ti_3C_2; and a new group of electrically conducting binders for use in Si anodes. Although results vary, many of these projects have achieved relatively stable cycling, showing over 1,000 cycles (Figure 3) in a ½ cell configuration, over 1,000 mAh/g capacity, and the ICL has been reduced to under 15%. The next phase of this work is to tackle the remaining issues of SEI stability and higher loadings that will make the electrode structures more relevant to commercial batteries. Testing will also be migrated to full cells.

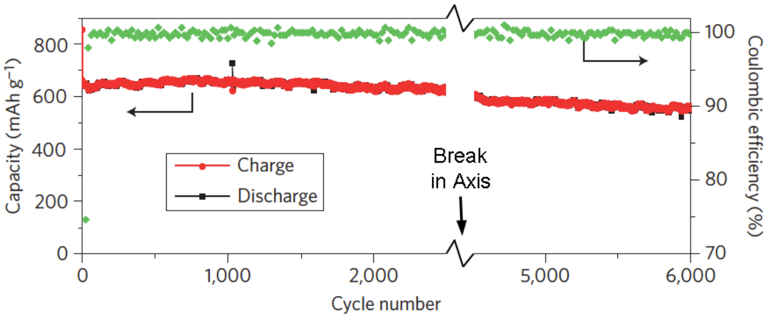


Figure 3. 6,000 Cycles of a 1/2 Cell Containing Si Nanotubes (Stanford University)

Researchers are also developing and testing other alloy based anodes. One, $SiO\text{-}Sn_{30}Co_{30}C_{40}$, delivers over 1,000 mAh/g with ~20% ICL and negligible capacity fade over 100 cycles in a half cell. Recently, Argonne announced the licensing of a Si/graphene composite material to CalBattery,[1] which has reported that independent tests in full Li-ion cells showed (with advanced cathode and electrolyte materials) 525 Wh/kg and specific anode capacity of 1,250 mAh/g.

New and Improved Alloy Anodes

The main "next generation" anode technologies to be pursued will be alloy based, predominantly silicon and tin based anodes. Nano sizing will continue to be pursued, utilizing both "traditional" approaches (Si nano particles or nano-wires grown on traditional graphite or graphene particles for standard electrode processing), and more novel approaches such as Si nanowires grown directly on the current collector, or Si and Sn multi-metal alloys. Particular attention will be paid to SEI stability on cycling, to the ICL, and to particle stability, which is sometimes compromised by the large volume change experienced by these materials on cycling. The large volume change on cycling can easily render Si and other alloy anodes no better than graphite on a volumetric capacity basis. The goal is to increase the amount of energy that can be stored in a given volume (and to reduce the cost of that energy storage). Thus, volumetric capacity is a critical parameter and each of these materials and approaches must demonstrate a significant advantage against graphitic carbons.

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[1] http://www.anl.gov/articles/argonne-and-calbattery-strike-deal-silicon-graphene-anode-material.

On a more exploratory front, some research into conversion reaction materials (e.g., CoO, Fe_2O_3, and CuF) may be undertaken. These materials provide large capacities, often more than 600 mAh/g and very high volumetric capacities. However, the issues with these materials are numerous including: poor kinetics; poor capacity retention on cycling (often due to metal agglomeration); large irreversible capacity loss; and large voltage hysteresis.

Advanced Electrolytes

Current electrolytes, which typically include a blend of cyclic and linear carbonate solvents and $LiPF_6$ salt, provide good performance and stability. However, the solvents are highly flammable and typically have a high vapor pressure, which causes them to out gas at elevated temperatures, building up pressure within cells over time. Also, the $LiPF_6$ salt is known to react almost instantly with water, producing HF, which in turn attacks nearly all elements of the cell. This reaction, along with the instability of $LiPF_6$ above ~80°C, leads in part to the challenges in Li-ion cells' high temperature capability.

Work on new electrolytes and additives is focused on one or more of the possible improvement areas of high voltage stability; high temperature stability, low temperature operation; abuse tolerance; lower cost; and possibly longer life through SEI stabilization. The exploratory materials program is supporting seven electrolyte projects which are developing plastic-like glassy electrolytes; flame retardant liquid electrolytes; single ion conductor electrolytes (which would enable the use of much thicker electrodes); new salts providing better high temperature stability; and electrolytes that enable much lower temperature operation, Figure 4. In addition, the program is supporting the theoretical investigation into high voltage stability and electrolyte blends that may lead to more stable SEIs on graphite. Researchers are also developing electrolytes with improved high voltage stability; phosphazene based electrolytes that promise improved abuse tolerance; and recently announced the availability for license of an overcharge shuttle for Li iron phosphate cells.

Nearly every development program includes an electrolyte improvement effort. Most are focused on enabling higher voltage operation either to support the Li-Mn rich oxide cathodes or the use of traditional NMC at higher voltage. Several have developed electrolyte blends or additives to stabilize the SEI on Si containing anodes. Finally, one developer is perfecting the use of LiTFSI salt that offers the possibility of much higher temperature stability and therefore longer battery life.

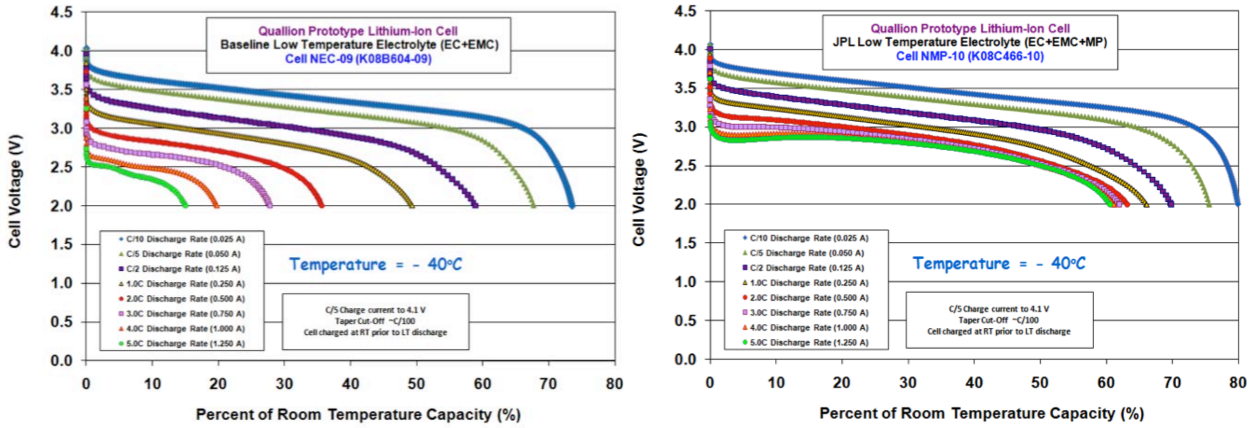

Figure 4. Discharge Capacity for a Baseline Electrolyte (left) and an Improved Methyl Proprionate Electrolyte (right) for Cells Cycled from C/10 to 5C

New Electrolytes

Work will continue on new flame retardant electrolyte additives, new inflammable solvents, and new salts that offer improved high temperature stability. In addition, specific additives will be sought to help stabilize the SEI on alloy anodes, and to stabilize the surface of high voltage cathodes like $LiMn_{1.5}Ni_{0.5}O_4$. On a more exploratory front, solid and gel electrolytes will continue to developed. Where appropriate, ionic liquids will be evaluated for their promise of enhanced abuse tolerance and stability.

Separators

Current work is focusing on developing separators that provide enhanced abuse tolerance, better high voltage stability, and improved low temperature operation. Some of the technologies being developed include a ceramic impregnated separator that shows much improved low temperature performance and greatly increased high temperature melt integrity. The latter may be important to the avoidance of shorts during high temperature excursions that can occur when traditional separators shrink. Another is developing a separator and process to permit direct deposition onto anode and/or cathode sheets.

Manufacturing Innovations

Manufacturing costs can be a significant fraction of cell and system costs, Figure 5 shows results from BatPAC from Argonne. Thus DOE and U.S. DRIVE are investigating manufacturing techniques that have potential to increase cell performance while reducing cost, including: new UV and EV curable binders to permit much faster and less expensive slurry drying; use of aqueous or dry binding technologies; and fast formation techniques. In the laboratory programs, researchers are investigating technologies to produce very thick (1 mm vs. 100 µm) electrodes with aligned pores; spray pyrolysis techniques for active material production; and new diagnostic technologies to investigate manufacturing techniques in-situ.

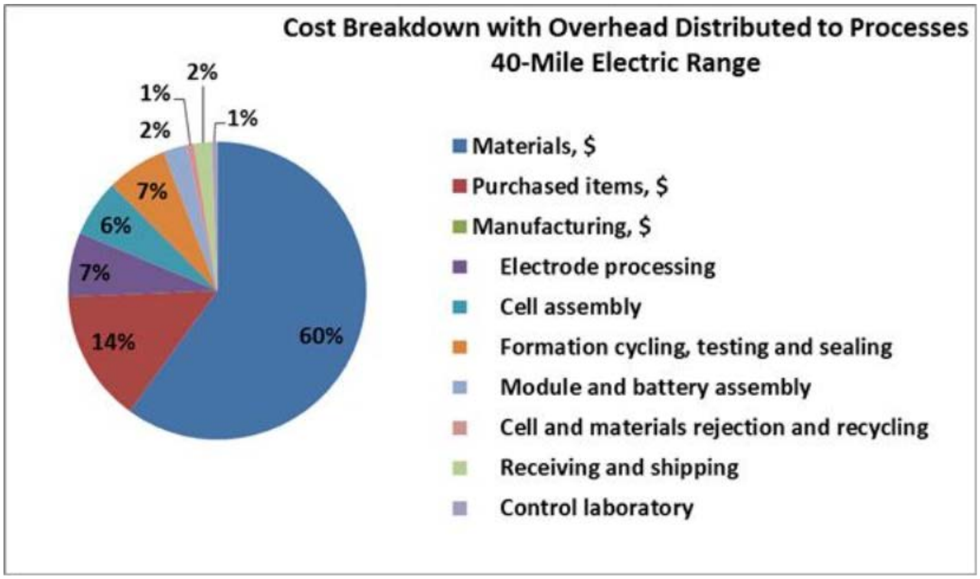

Figure 5. Cost Breakdown of PHEV40 Battery (manufacturing related costs amount to 25% of the battery, plus approximately 50% of the material cost)

Enhanced Abuse Tolerance

The design of abuse tolerant energy storage systems begins with the specification of relevant abuse conditions and the desired responses to those conditions. USABC-sponsored development programs include characterizations of the candidate technologies in abuse tests. Uniform standards of characterization testing in this area have been established, and abuse test manuals are available at http://www.uscar.org/guest/article_view.php?articles_id=86. The advanced material and cell programs fund projects to improve the intrinsic stability of Li-ion battery chemistries through development of new materials, and characterization of advanced commercial materials. Many of the research directions listed above have direct impacts on improved abuse tolerance. Some of those include coated cathodes and anodes, non-flammable electrolytes, solid polymer and glassy electrolytes, ceramic coated or impregnated separators, and overcharge shuttles and polymer overcharge protection materials. For example, Argonne and SNL are evaluating an AlF_3 cathode coating that significantly slows down the cathode thermal runaway process, as shown in Figure 6. Researchers are also evaluating polymer materials that conduct electricity above a certain potential, thus providing an overcharge protection mechanism. An overcharge shuttle appropriate for Li iron phosphate batteries has been developed and licensed by Argonne. Coatings and concentration gradient cathode materials are also being developed with the goal of enabling higher voltage operation and of enhancing the abuse tolerance of Li-ion batteries. Finally, phosphazene based electrolytes are being developed at INL and tested at SNL and are showing promise in reducing the heat released during thermal runaway. Developers have developed or are developing: a heat resistant layer (added to one or both of the electrodes) to enhance the cells' ability to avoid internal shorts; coated and ceramic impregnated separators to guard against internal short circuits and to provide enhance high temperature melt integrity (HTMI); and novel thermal management technologies to more closely control the temperatures that cells are exposed to.

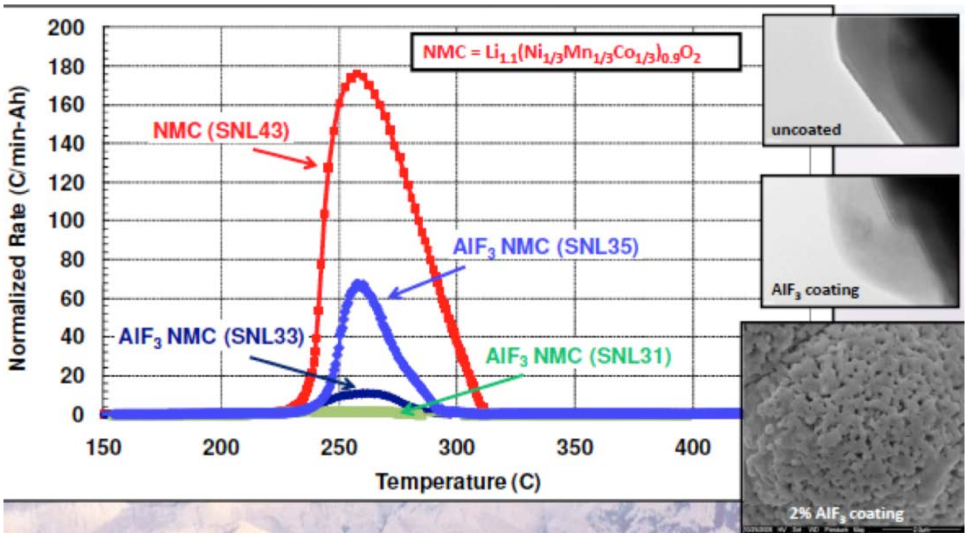

Figure 6. AlF_3 Coating Improves the Thermal Stability of NMC Materials by Increasing the Thermal Runaway Temperature by 20°C and Reducing the Peak Heating Rate

Additional activities may include: preparing a "Permanent SEI"; new separators (and/or new ceramic coatings applied to separator or electrode); investigating the abuse tolerance of batteries containing advanced anodes (made with Si or other alloys) and advanced cathodes (such as the Li-Mn rich oxide cathodes); new materials R&D for enhanced thermal abuse tolerance, e.g., high thermal conductivity materials for use in cell design.

Improved Thermal Management

Thermal management is critical to achieving system life and abuse tolerance. Current thermal management technologies add weight, cost, and complexity to the energy storage system. The use of novel thermal management approaches could both manage the battery's temperature and potentially reduce overall cost. Approaches that significantly extend the upper or lower operating temperature ranges of the system are also of interest. DOE may investigate novel materials that could be integrated into cell designs for faster heat rejection and infusion.

Computer Aided Battery Design (CAEBAT)

DOE is supporting the development of Computer Aided Battery Design software with the goal of developing an integrated suite of battery design software tools. Electrochemical performance simulations and thermal design software are being improved and integrated to form a full battery design suite. The process of testing new materials in multiple cell sizes, in multiple battery pack designs, and over many months is extremely time consuming and expensive. This software suite could greatly speed the design of new batteries and provide critical guidance to developers.

A final project that is related to the above involves a Materials Project web site and database. The web site provides free searchable access to general materials properties covering over 15,000 inorganic compounds with the number of compounds increases continuously. The site contains tools ("apps") designed to aid in materials design for specific application areas such as Li-ion battery technology.

Beyond Li-Ion Battery R&D

The "beyond Li-ion" technologies, such as Li/sulfur, and Li/air, among others, offer a further increase in energy and potentially greater reductions in $/Wh compared to next-gen lithium ion batteries. However, these systems require many more breakthroughs, some on a fundamental material level, before they can be considered for real-world use. The work on these systems still spans the entire program area. Samples of that work are provided below; more complete descriptions are available in the VTP annual progress reports available at http://www1.eere.energy.gov/vehiclesandfuels/resources/fcvt_reports.html.

DOE is investigating the fundamental issues associated with cycling Li metal anodes as well as potential solutions to those issues. The main research topics under investigation include: coatings, novel oxide and sulfide-based glassy electrolytes, and in-situ diagnostics approaches to characterize and understand Li metal behavior during electrochemical cycling.

Researchers in the advanced materials R&D program are developing two separate electrolytes for Li/air systems; investigating the role of catalysts on Li/air cathode reversibility and hysteresis; novel carbons for Li/air cathode applications; novel sulfur cathode architectures based on mesoporous carbons; and polysulfide solvents to manage polysulfide concentrations in the electrolyte. Researchers in the advanced cell R&D program are also developing and testing a series of organosilicon electrolytes in Li air cells. Work by developers is focused on commercializing a block copolymer electrolyte that impedes Li dendrite formation (this technology has shown thousands of cycles with little capacity degradation, and has also shown good abuse tolerance through testing by independent third parties). Other work is progressing on a nanocomposite sulfur cathode (with accompanying electrolyte), Figure 7; and on a silane based electrolyte for use in Li/S cells.

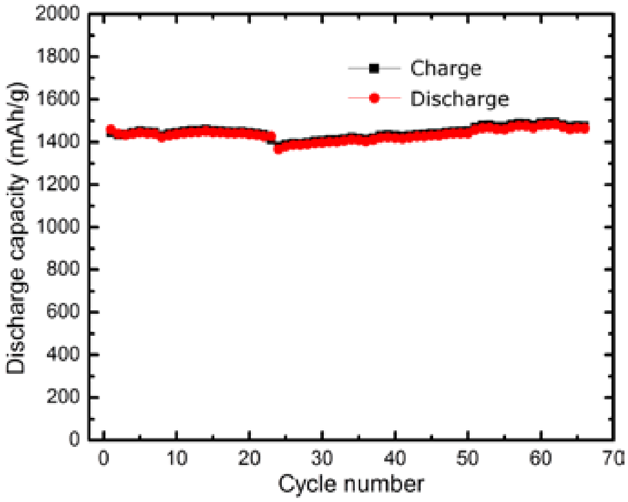

Figure 7. Performance of a Li/S Cell with a New Electrolyte Developed by the Team of Penn State University, EC Power, Johnson Controls Inc., and Argonne National Laboratory

Possible New Areas of Beyond Li Ion Battery R&D

The challenges facing beyond lithium-ion battery systems are numerous. Issues remain to be solved on the cathode (e.g., stabilizing or containing the lithium polysulfides or developing air cathodes with much smaller hysteresis, better rate, and better reversibility), the anode (stabilizing the lithium metal interface, combatting the creation of mossy lithium that reacts with electrolyte and "dries out" the cell), and the electrolyte (flammability of liquids, stability of liquids against lithium air reaction products, creating a solid electrolyte with good rate and mechanical properties, etc.). In addition to these technical challenges, there is continuing uncertainty about the ability of lithium air and lithium sulfur batteries to meet the volume and cost goals of automotive batteries for PEVs. Some of the research that will be pursued in coming years includes:

- Continue to support efforts to stabilize the lithium metal interface during cycling. Options to be evaluated include coatings, dopants, solid glassy electrolytes, electrolyte dopants, and others.
- Expand and evaluate options for stabilizing the sulfur cathode. Recent attempts in the literature include core/shell like approaches and egg/yolk structures to isolate the polysulfides from direct contact with the electrolyte, the use of mesoporous carbon to slow the dissolution of polysulfides, and search for solvents to remove lithium sulfides from the anode interface.
- A more fundamental investigation of the reaction mechanisms and dynamics on the air cathode are needed. U.S. DRIVE will work closely with the recently awarded Energy Storage Hub team to further this type of investigation.
- Study the impact of carbon structure and pore distribution on air cathode performance. Reaction products at the air cathode can "choke off" further reactions. The surface area and pore distribution have been shown to impact the utilization of the air cathode.
- Investigate low cost catalysts for air cathodes. Some researchers have found greatly increased activity on OER and ORR reactions using catalysts. Unfortunately, to date those catalysts have been extremely expensive materials, like gold and platinum. A fundamental understanding of the role of those materials is needed in order to consider replacement materials.
- New electrolytes for air and sulfur batteries may be needed due to the continued observation of poor stability against Li metal and air cathodes in Li air cells, and due to the issue of polysulfide dissolution in Li sulfur cells.
- Also, as with Li-ion cells, the use of highly volatile liquid electrolytes may introduce abuse tolerance issues that will have to be addressed.

In addition to the specific technical topics listed above, multi-valent materials, like Mg, may be investigated along with other non-Li systems like Na, Zn, or Al.

Schedule and Milestones

The U.S. DRIVE energy storage R&D effort is focused on overcoming the remaining technical and cost barriers to commercialization of PEV batteries. Table 4 shows the high-level milestones in the energy storage area over the next several years.

Table 4. Overview of Out-Year Focus Areas for Energy Storage R&D

ROADMAP	2014	2015	2016	2017	2018	2019	2020
Goals	PHEV40						EV200
Cost ($/kWh)	300						125
Pack-specific Energy (Wh/kg)	100						200
Cell-specific Energy (Wh/kg)	200						300
Anode Capacity (mAh/g)	300						1,200
Cathode Capacity (mAh/g)	220						800
Battery Development Advanced electric drive vehicle battery development focused on cost reduction, life improvement, and abuse tolerance	1 5	7 8 MM-1		12 13	16	17	MM-2
Advanced Cell R&D Research focused on improving energy, power, and durability of promising electrochemical systems	2 6		9 11		15		
Advanced Materials Research focused on higher power, higher energy, and longer lived materials	3 4	10		14			

1. Initiate development of computer aided engineering tools to enable safer and more durable battery designs.
2. Increase support of materials production processes for high capacity cathodes with the potential to reduce battery material costs.
3. Initiate R&D awards to develop lower cost, high voltage, and non-flammable electrolytes.
4. Organize new national lab and university focus group to mitigate issues with alloy anodes.
5. Complete the 2nd Gen EDV Battery development contracts using higher capacity cathodes focused on the 2014 cost target of $300/kWh.
6. Increase support for 3rd Gen, Li Ion Materials & Cell R&D focused on advanced metal alloy or Si composite anodes.
7. Award 3rd Gen Li Ion PEV Battery Development contracts using safe, high voltage electrolytes and metal alloy or Si anodes focused on 2020 targets.
8. Begin commercialization of computer aided engineering tools to enable safer and more durable battery designs.
9. Initiate support to develop lower cost production for advanced metal alloy or Si anodes.
10. Initiate fundamental research focused on Li Metal and Beyond Li Battery technologies.
11. 2nd Phase R&D awards to develop lower cost, high voltage, and non-flammable electrolytes.
12. Test new overcharge shuttles, non-flammable electrolytes, and solid polymer electrolytes for enhanced abuse tolerance.
13. Complete the development of the CAEBATT Design Tools.
14. Increase support for Li Metal and Beyond Li Battery Materials Research.
15. Initiate R&D to develop durable and low cost Beyond Li Battery cells.
16. Award Next Gen Li Ion PEV Battery Development contracts using high voltage electrolytes and metal alloy or Si anodes focused on 2020 targets.
17. Award Beyond Lithium Ion PEV Battery Development contracts focused on 2022 targets.

Major Milestones

MM-1 Demonstrate through cell performance testing and detailed cost modeling batteries that can achieve the 2015 PHEV40 cost and performance goals.

MM-2 Demonstrate through cell performance testing and detailed cost modeling batteries that can achieve the 2020 EV200 cost and performance goals.