I. INTRODUCTION

Vehicle Technologies Program Overview

Energy Storage Research & Development Overview

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In 2011, sales of U.S. light duty vehicles rebounded slightly to approximately 12.8 million from less than 10 million in 2009. Sales of hybrid electric vehicles (HEVs) remain in the two to three percent range. The U.S. government continued its strong R&D support of electric drive vehicles (EDVs), including HEVs, plug-in hybrid electric vehicles (PHEVs), and pure electric vehicles (EVs). In 2011, additional facilities for advanced battery and battery component manufacturing, supported by the U.S. Department of Energy (DOE) under American Recovery and Reinvestment Act (ARRA) cost-shared grants[1], finished construction and began production. A description of the battery manufacturing grants is presented in Chapter II.

An important step for the electrification of the nation's personal transportation and for the continued success of the new domestic Li-ion battery manufacturing factories is the development of more cost-effective, long lasting, and abuse-tolerant Li-ion batteries. DOE's continuing R&D into advanced batteries for transportation offers the possibility of reducing our dependence on foreign oil and the negative economic impacts of crude oil price fluctuations. It also supports the Administration's goal of deploying 1 million PHEVs by 2015. During the fiscal year (FY) 2011, battery R&D work continued its focus on high-energy batteries for PHEVs and EVs.

I.A Vehicle Technologies Program Overview

The DOE's Vehicle Technologies (VT) Program develops advanced transportation technologies that would reduce the nation's use of imported oil. Technologies being supported by VT include hybrid drive technologies, advanced energy storage devices (batteries and ultracapacitors), power electronics and motors, advanced structural materials, and advanced combustion engines and fuels[2]. DOE works with the U.S. automakers through the United States Council for Automotive Research (USCAR)—an umbrella organization for collaborative research among Chrysler LLC, Ford Motor Company, and General Motors Company[3]. Collaboration with automakers through the US DRIVE Partnership enhances both the relevance and the potential for success of these programs. This partnership is focused on funding high-reward/high-risk research at national laboratories, universities, and with industry, that promises improvements in critical components needed for more fuel efficient and cleaner vehicles.

I.B Energy Storage Research & Development Overview

I.B.1 Programmatic Structure

The objective of the energy storage research and development effort within the VT Program is to advance the development of batteries to enable a large market penetration of hybrid and electric vehicles. Program targets focus on overcoming technical barriers to enable market success including: (1) significantly reducing battery cost, (2) increasing battery performance (power, energy, durability), (3) reducing battery weight & volume, and (4) increasing battery tolerance to abusive conditions such as short circuit, overcharge, and crush.

The energy storage effort includes multiple activities, from focused fundamental materials research to battery cell and pack development and testing. The R&D activities involve either short-term directed research by commercial developers and national laboratories, or exploratory materials research, generally spearheaded by the national laboratories and universities. There are three major inter-related and complementary program elements, namely:

- Advanced Battery Development, System Analysis, and Testing
- Applied Battery Research (ABR)
- Focused Fundamental Materials Research, or Batteries for Advanced Transportation Technologies (BATT)

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[1] http://www.whitehouse.gov/the-press-office/24-billion-grants-accelerate-manufacturing-and-deployment-next-generation-us-batter

[2] See http://www1.eere.energy.gov/vehiclesandfuels/ for more information.

[3] For more information, please see http://www.uscar.org/guest/view_partnership.php?partnership_id=1.

The *Advanced Battery Development, System Analysis, and Testing program*'s goal is to support the development of a U.S. domestic advanced battery industry whose products can meet electric drive vehicle performance targets. R&D in this activity focuses on the development of robust battery cells and modules to significantly reduce battery cost, increase life, and improve performance. The activity takes place in close partnership with the automotive industry, through our cooperative agreement with the United States Advanced Battery Consortium (USABC). In FY 2011, the USABC supported 12 cost-shared contracts with developers to further the development of batteries for EVs, PHEVs, and HEVs. DOE also works directly with industry battery and material suppliers via National Energy Technology Laboratory (NETL) contracts. In FY2011, NETL managed twenty one battery R&D contracts. Benchmark testing of emerging technologies is performed to remain abreast of the latest industry developments. Battery technologies are evaluated according to USABC Battery Test Procedures Manuals for the relevant EDV applications[4,5,6]. Additional R&D involves the development of innovative technologies to reduce the cost of thermal management systems, enhance battery tolerance to abusive conditions, and the development of computer aided engineering tools to enable battery design optimization.

The *Applied Battery Research* (ABR) activity is focused on the development of next generation, high-energy lithium-ion battery electrochemical couples that have the potential to meet US DRIVE battery-level targets for 40 mile all electric range PHEV batteries. The work is carried out by a team headed by the Argonne National Laboratory (ANL) and involves several other national labs, and universities. The emphasis of ABR is to overcome the major technical barriers related to long range PHEV batteries, including: (1) inadequate energy density and specific energy to meet the "charge-depleting" energy requirement, within the weight and volume constraints, for the 40-mile all-electric-range mid-size passenger PHEV, and (2) insufficient cycle life stability to achieve the 3,000 to 5,000 "charge-depleting" deep discharge cycles.

The *Focused Fundamental Research* activity, also called the Batteries for Advanced Transportation Technologies (BATT) activity, addresses fundamental issues of chemistries and materials associated with lithium batteries. It attempts to gain insight into system failures, develops models to predict failure and to optimize systems, and researches new and promising materials. It emphasizes the identification and mitigation of failure modes, coupled with materials synthesis and evaluation, advanced diagnostics, and improved electrochemical models. Battery chemistries are monitored continuously with periodic substitution of more promising components based on advice from within this activity, from outside experts, and from the assessments of world-wide battery R&D. The work is carried out by a team headed by the Lawrence Berkeley National Laboratory (LBNL) and involves several other national labs, universities, and commercial entities.

Several Small business innovation research (SBIR) contracts are also supported by VT, in addition to the R&D described above. SBIR projects have been the source of new ideas and concepts over the years. These SBIR projects are focused on the development of new battery materials and components.

Coordination within DOE and with other government agencies is a key attribute of the VT energy storage R&D effort. VT coordinates efforts on energy storage R&D with the DOE Office of Science, the DOE Office of Electricity, and the Advanced Research Projects Agency – Energy (ARPA-E). VT also has established extensive and comprehensive ongoing coordination efforts with other government agencies in energy storage R&D. Such efforts include membership and participation in the Chemical Working Group of the Interagency Advanced Power Group (IAPG), active participation in program reviews and technical meetings sponsored by other government agencies, and coordinating the participation of representatives from other government agencies in the contract and program reviews of DOE-sponsored efforts DOE also coordinates with the Department of Transportation/National Highway Traffic Safety Administration (DOT/NHTSA), the Environmental Protection agency (EPA), and with the United Nations Working Group on Battery Shipment Requirements. Additional international collaboration occurs through a variety of programs and initiatives. These include: the International Energy Agency's (IEA's) Implementing Agreement on Hybrid Electric Vehicles (IA-HEV), the eight-nation Electric Vehicle Initiative (EVI), and the Clean Energy Research Center (CERC) bilateral agreement between the US and China.

[4] United States Advanced Batteries Consortium, USABC Electric Vehicle Battery Test Procedure Manual, Rev. 2, U.S. Department of Energy, DOE/ID 10479, January 1996.

[5] U.S. Department of Energy, PNGV Battery Test Procedures Manual, Rev. 2, August 1999, DOE/ID-10597.

[6] United States Council for Automotive Research, RFP and Goals for Advanced Battery Development for Plug-in Electric Vehicles, http://www.uscar.org/.

I.B.2 Some Recent Highlights

This section contains brief summaries of some key technical accomplishments in FY 2011 resulting from the Energy Storage R&D and associated efforts. These accomplishments were selected from the many active projects and each represents a significant degree of accomplishment within the project, or the completion of a significant milestone, or a significant breakthrough of another kind that took place during the year.

Electric Drive Vehicle Market

- **Significant US sales of electric vehicles began.** GM's Chevrolet Volt EREV, using technology developed by DOE and licensed to GM, achieved 2011 sales of 7,671 vehicles. Sales of the Nissan Leaf EV were 9,674 in 2011. (Both the Volt and the Leaf sold more in 2011, their first year in the market, than did the 2000 Toyota Prius, which sold 5,562, or the first hybrid in the market, the Honda Insight, which sold 3,788.)
- **DOE-supported technologies continue to move into commercial applications.** Several technologies, developed partially under VT-sponsored projects, have moved into commercial applications. Hybrid electric vehicles on the market from BMW and Mercedes are using lithium-ion technology developed under projects with Johnson Controls–Saft (JCS). Lithium-ion battery technology developed partially with DOE funding of a USABC project at LG Chem is being used in GM's Chevrolet Volt extended-range electric vehicle and has been selected for the upcoming Ford Focus EV battery. LG Chem will also supply Li-ion batteries to Eaton for hybrid drive heavy vehicles. Johnson Controls-Saft continued to supply Li-ion battery packs to Azure Dynamics for electric delivery vans built on the Ford Transit Connect platform. A123Systems is producing lithium-ion battery systems for the Fisker Karma EV and the Navistar Modec Electric trucks. A123Systems has been selected to supply lithium-ion batteries for use in the GM Spark EV, the BMW ActiveHybrid 5 and 7 models, and VIA Motors electric trucks.
- **Recovery Act Facilities Projects Initiated and Production Underway.** All projects for battery and materials manufacturing facilities funded by the American Recovery and Reinvestment Act were initiated. Production began at several facilities, including;
  - GM Battery pack assembly facility in Brownstown, MI
  - A123Systems Cell and pack assembly at Livonia, MI
  - Johnson Controls cell and pack assembly in Holland, MI
  - Saft cell and pack assembly in Jacksonville, FL
  - Enerdel cell and pack assembly in Indianapolis, IN
  - East Penn advanced lead acid battery plant in Lyon Station, PA
  - Celgard separator material production at Charlotte, NC
  - Toda cathode production plant in Battle Creek, MI
  - Pyrotek anode production plant in Sanborn, NY
  - Honeywell electrolyte salt pilot production facility in Buffalo, NY

Batteries – High Energy

- **EV cells exceed USABC EV targets.** K2 Energy Solutions' new 'flat pack' 45-Ah energy cells, based on $LiFePO_4$ technology, exceed USABC EV energy density targets of 230 Wh/L and 150 Wh/kg.
- **PHEV cell energy density and pack volumetric efficiency improved.** LG/CPI demonstrated prototype lithium-ion cells using advanced Mn-rich layered-layered composite cathode materials, with resulting cell energy density increase of 20% over LG Chem's previous baseline chemistry large format PHEV cell, and has also designed and analyzed an automotive grade, self-contained battery pack using a refrigerant-based thermal management system, which reduces total pack volume by 20% over its previous PHEV pack design.
- **PHEV system-level energy density significant improved.** Johnson Controls developed a new PHEV prismatic cell/system technology that offers twice the all-electric range and a 15% reduction in system volume versus their baseline technology.
- **EV cell specific energy improved using NCM chemistries.** Based on data from prototype large format (0.357 L) lithium-ion cells using various NCM chemistries, Cobasys developed and demonstrated specific energy density

increase to greater than 160 Wh/kg (greater than 80% of the target specific energy of 200 Wh/kg) without compromising life, performance and abuse tolerance.

- **High-energy EV cells (215 Wh/Kg) using high-energy cathode.** Envia Systems scaled-up synthesis of high-capacity cathode material to 5 kg per batch, and fabricated and delivered twenty 20-Ah EV cells for testing at INL. Envia developed a new electrolyte composition that operates at high voltage, shows greater low-temperature conductivity, and exhibits similar cycle life and power characteristics when compared to Envia's current baseline electrolyte.
- **Thick electrode designs for ultrahigh energy density**. Massachusetts Institute of Technology developed a new electrode construction technique for thicker (10 times) electrodes. The approach is based on a sintered electrode architecture with aligned, low-tortuosity porosity. In collaboration with LBNL, a freeze-casting process was developed which results in directional solidification of ice with desired pore morphologies and rejects solid particles from the growth front. Freezing rate and hygroscopic additives alter the microstructural morphology. Freeze-cast $LiCoO_2$ cathodes, with aligned porosity and a thickness of 220 microns, showed good high-rate capability and achieved >120 mAh/g at the 2C rate.
- **High-energy Si anodes suitable for large format lithium-ion batteries.** National Renewable Energy Laboratory developed a technique for hot wire chemical vapor deposition of amorphous-Si powders to make amorphous-Si electrodes of sufficient thickness for high energy batteries. Electrodes 15-µm thick displayed an initial capacity of ~2500 mAh/g, with a durable capacity of ~1,000 mAh/g (about 3 times that of graphite).
- **Ion-exchanged cathode material shows improved rate capability and high energy.** Argonne National Laboratory developed a sodium/lithium ion-exchange process to produce a new high-energy cathode material with reduced site disorder and improved rate capability. From precursors of layered $Na[M]O_2$ metal oxides, an ion-exchanged $Li[M]O_2$ layered oxide was produced with a composition of $Li_{1.32}Na_{0.02}Ni_{0.25}Mn_{0.75}O_x$. This material demonstrated >220 mAh/g at 4.8 V over 40 cycles, compared to current cathode materials providing ~150mAh/g at 3.7 V.

Batteries – Improved Power

- **EV battery power improvement.** Quallion LLC, which is developing a hybrid battery with separate high power and high energy modules, demonstrated a 30% higher specific power at 80% DOD discharge than a comparable battery without the high power module.
- **Power cells with good cyclability and low-temperature performance.** ActaCell developed and built 8-Ah pouch cells with their stabilized manganese spinel cathode; these cells are expected to achieve USABC targets for cycle life and low-temperature performance.
- **Multifunctional inorganic-filled separator for large format Li-ion batteries.** Entek Membranes demonstrated improved thermal stability, cycle life, and power capability in 18650 cells with this separator, and affirmed the potential to meet USABC's cost target in scaled-up production.
- **Power improvement for high-energy layered-layered cathodes**. Oak Ridge National Laboratory demonstrated that a nm-thick coating of lithium phosphorus oxynitride (Lipon) on the high-energy Li-rich or layered/layered cathodes dramatically improves rate capability, one of the critical needs for commercialization. The uncoated material's capacity dropped by 50% when the discharge rate is increased from C/5 to 2C, while cells with the Lipon-coated cathodes retained over 80% capacity. The Lipon-coated sample also demonstrates respectable capacity (120 mAh/g) even at the 15C rate.

Batteries – Improved Life and Abuse Tolerance

- **PHEV cell calendar life improvement.** Based on analysis of results from a multi-temperature matrix of long-term calendar life test data, A123Systems' developed 19.6-Ah prismatic PHEV cells are projected to exceed the USABC 15 year Calendar Life goal.
- **EV cells demonstrate excellent cycle life.** SK Innovation's production-ready 25-Ah high-energy EV cells show cycle life that is double the USABC target, with good safety.
- **Determination of Aging Path Dependence in Batteries.** Idaho National Laboratory and the University of Hawaii developed tools to better understand the mechanisms of cell aging. Aging mechanisms are very sensitive to environmental and duty-cycle conditions. The tools use application-relevant testing, accurate diagnostic techniques,

and physics-based life models. The results can revolutionize battery management, and enable a realistic solution to operate battery systems reliably and safely under highly diverse usage scenarios.

- **LiF anion binding agent electrolytes for enhanced abuse tolerance.** Sandia National Laboratory and Binrad Industries obtained dramatic improvements in the thermal stability of cathodes and improvements in cell runaway response using electrolytes with LiF/anion binding agent salts. The specific heat measured for a NMC433 cathode was much less in LiF/ABA (611 J/g) than in $LiPF_6$ (1132 J/g).

Ultracapacitors

- **Prismatic Li-ion ultracapacitors with dry film electrodes.** Maxwell Technologies increased the form factor of their stacked lithium-ion capacitor cells from 35 F to 250 F using their dry film electrode build process, demonstrated low temperature performance at -30°C, and utilized full format, prismatically wound cells as the foundation of a refined system architecture that is the basis for low cost prototype systems to be constructed and delivered in 2012 with a 20% reduction in size and 17% reduction in cost over the previous iteration.
- **New ultracapacitors for power-assist HEVs.** Maxwell Technologies developed an asymmetric capacitor with greatly increased energy density address USABC's low-energy energy storage system (LEESS) requirements. Maxwell successfully applied their proprietary dry process electrodes to produce small laboratory-sized cells that can be cycled at higher voltages than current ultracapacitors, further increasing the energy density. Maxwell is also developing a new system for manufacturing the cells to minimize cost and approach the USABC target for selling price.

New Technologies

- **High-voltage Single-Ion Conducting electrolytes.** Lawrence Berkeley National Laboratory developed a new Single-Ion Conductor (SIC) gel, based on a polysulfone/carbonate blend, that may enable thick electrodes. The impedance of the polysulfone-TFSI gel against lithium metal is an order of magnitude less than the base material, and the interfacial impedance is now very close to values enabling its use in a practical cell. When used as a binder in composite electrodes, the PS-TFSI SIC gel was shown to be stable up to at least 4.5 V, suitable for use with several high energy cathodes, including the spinel–type and composite cathodes.
- **$LiCoPO_4$ cathode and high-voltage electrolyte.** Army Research Laboratory (ARL) found a new electrolyte additive to significantly improve high-voltage stability. The additive, tris(hexafluoroisopropyl)phosphate, a highly fluorinated phosphate ester with a fluorine/hydrogen ratio of 6, enhances the high-voltage stability of carbonate electrolytes. ARL also developed a 4.8-V cathode material, Fe-$LiCoPO_4$, in which a portion of the Co is substituted by Fe^{2+} and Fe^{3+}, which showed improved stability and cycle life compared with $LiCoPO_4$.
- **High-voltage spinel cathode performance improved via doping.** Pacific Northwest National Laboratory developed a chromium-doped high-voltage spinel, $LiNi_{0.45}Cr_{0.05}Mn_{1.5}O_4$ that exhibited stable cycling and greatly improved efficiency. PNNL also found that low concentrations of Li bis(oxalato)borate (LiBOB) not only improves the first-cycle efficiency of $LiNi_{0.45}Cr_{0.05}Mn_{1.5}O_4$ from 76% to 85%, but the rate capability also increases. LiBOB is believed to form a protection film on the cathode surface which is resistive to the damaging HF and POF_3 generated by reactions of the $LiPF_6$ salt in the electrolyte.
- **Silicon-based anode exceeds targets.** 3M developed new class high-energy anode materials, demonstrated manufacturability, and optimized electrode/cell design to achieve high cycle-life. These materials are now being commercialized. The design of the alloy was based on the active/inactive alloy concept, and achieved a reversible capacity of 1500 mAh/cc after full lithiation and expansion. At this capacity the alloy is expected to have a volumetric expansion of 100% during lithiation and increase the energy density of a lithium ion cell by 15-20%, depending on the cathode formulation.

New Tools and Techniques

- **Materials search engine with lithium electrode explorer.** Lawrence Berkeley National Laboratory and Massachusetts Institute of Technology launched the first Google-like materials search engine. The web site contains a database of over 15,000 computed compounds for general searches and a Li-battery electrode explorer ('apps') designed to aid in materials design for specific applications.
- ***In situ* X-ray diffraction for diagnostics of cathode materials.** Brookhaven National Laboratory used synchrotron-based *in situ* X-ray diffraction to study the structural changes of cathode materials of Li-ion batteries during chemical

lithium extraction. This technique enables evaluations of critical nanometer-scaled microstructural and micro-chemical changes as a function of battery test conditions, electrode materials, electrolyte, and electrolyte additives. The new technique was used to investigate the phase transitions of $LiFePO_4$.

- **Multi-Scale Multi-Dimensional (MSMD) framework for battery design.** As part of DOE's Computer-Aided Engineering for Electric Drive Vehicle Batteries (CAEBAT) program, National Renewable Energy Laboratory (NREL) developed and published a methodology for multi-domain modeling of lithium-ion batteries encompassing multi-physics in varied length scales. NREL's MSMD framework introduces multiple computational domains for corresponding length scale physics, and decouples geometries between submodel domains while coupling physics bi-directionally.
- **Battery cost model peer-reviewed and published.** The Battery Performance and Cost model (BatPaC v1.0) developed by Argonne National Laboratory has been made publically available, posted on the web along with supporting documentation. This version of the model includes significant improvements in response to multiple peer-reviews. An extensive formal peer review was conducted by an independent panel commissioned by the EPA.

I.B.3 Organization of this Report

This report covers all the projects currently ongoing or starting as part of the energy storage R&D effort within the Office of Vehicle Technologies. Chapter II contains information on the projects which are funded under the American Recovery and Reconstruction Act (ARRA) of 2009 (the Recovery Act). Chapter III focuses on the battery development program. Chapter IV lists all the projects which are being conducted under the Applied Battery Research activity in which ANL has a leading role. Similarly, Chapter V lists all the projects which are part of the Focused Fundamental Research activity with a leading role by LBNL. A list of the individuals who contributed to this annual progress report or otherwise are collaborating with the energy storage R&D effort appears in Appendix A. A list of acronyms is provided in Appendix B. An electronic version of this report can be accessed at http://www1.eere.energy.gov/vehiclesandfuels/resources/fcvt_reports.html.

We are pleased with the progress made during the year and look forward to continued work with our industrial, government, and scientific partners to overcome the remaining challenges to delivering advanced energy storage systems for vehicle applications.

David Howell
Team Lead, Hybrid and Electric Systems
Vehicle Technologies Program

Tien Q. Duong
Manager, Exploratory Technology Research
Vehicle Technologies Program

Peter W. Faguy
Manager, Applied Battery Research
Vehicle Technologies Program

Brian Cunningham
Lead, Battery Testing, Analysis and Design
Vehicle Technologies Program