FY 2002 Status Report on the U.S. Department of Energy Electric and Hybrid Electric Vehicle Energy Storage Technologies Research

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Abstract

The U.S. Department of Energy (DOE), through its FreedomCAR and Vehicle Technologies Program, supports active, long-range research and development (R&D) to develop and commercialize electric vehicle (EV) and hybrid electric vehicle (HEV) technologies. This research is conducted in partnership with DOE's national laboratories, the auto industry/suppliers, other government agencies, universities, and small businesses. This paper summarizes the fiscal year (FY) 2002 status and the prior year's primary accomplishments for R&D programs to develop high-energy batteries for electric vehicles and high-power batteries for hybrid electric vehicles. The EV Battery program focuses on the development of advanced lithium-based batteries, which is carried out by the U.S. Advanced Battery Consortium (USABC). The High Power Batteries program focuses on candidate battery chemistries (nickel metal hydride and lithium-based electrochemical couples) that have been identified as most likely to succeed in meeting the requirements for high-power batteries for use in HEVs. The Batteries for Advanced Transportation Technologies (BATT) program researches high-performance rechargeable batteries for use in EVs and HEVs and addresses fundamental issues of chemistries and materials that face all lithium battery candidates for such applications. The Advanced Technology Development (ATD) program at DOE's national laboratories assists industrial developers of lithium-ion batteries in development of a low-cost, long-life, safe, and high-power energy storage device that meets or exceeds specific requirements for power-assist and dual-mode hybrid vehicles. This paper summarizes progress of the R&D projects toward improving the performance of battery components and materials and developing new and improved methods to characterize and monitor the performance of battery components. It summarizes the technology objectives, technical barriers, the approach for overcoming those barriers, recent accomplishments, current status, and future plans.

Keywords: Battery, electric vehicle, hybrid electric vehicle, lithium-ion, power.

1 Introduction

The successful commercialization of Electric Vehicles (EVs) and Hybrid Electric Vehicles (HEVs) directly contributes to reducing the United States' growing dependence on petroleum fuels for transportation; decreasing polluting and greenhouse gas emissions from vehicles; and facilitating a long-term transition to sustainable renewable energy sources. Recognizing these potential benefits, the United States actively supports R&D to develop EV and HEV technologies and to accelerate their commercialization. DOE's FreedomCAR and Vehicle Technologies Program plays a prominent role in this effort. FreedomCAR, a new government-industry program for the advancement of high-efficiency vehicles, focuses on fuel cells and hydrogen produced from renewable energy sources. It envisions affordable full-function cars and trucks that are free of imported oil and harmful emissions, without sacrificing safety, freedom of mobility, or the freedom of vehicle choice. The Program supports the development of advanced energy storage and power electronics technologies, fuel cells, advanced direct-injection engines, vehicle systems, lightweight materials, and fuels for transportation applications. Innovative research in these areas is supported through DOE's national laboratories, the auto industry and its suppliers, other government agencies, universities, and small businesses. The strategic approach for the Program includes:

- Develop technologies to enable mass production of affordable hydrogen-powered fuel cell vehicles and ensure the hydrogen infrastructure to support them.

- Continue support for other technologies to reduce oil consumption and environmental impacts.
- Develop technologies applicable across a wide range of passenger vehicles instead of single-vehicle goals.

The technology-specific 2010 goals of the DOE's FreedomCAR and Vehicle Technologies Program are listed in Table 1. As seen in Figure 1 for DOE's FY 2002 R&D budgets, the Program's energy storage efforts are focused on the high energy EV batteries, batteries for advanced transportation technologies (BATT), the high-power energy storage program, and the Advanced Technology Development (ATD) program. Each of these programs is discussed below.

Table 1: Technology-specific goals for the DOE's FreedomCAR and Vehicle Technologies Program[a, b]

Objective	Technology	Goals	
		Technical	Cost
Ensure reliable systems for future fuel cell power-trains with costs comparable with conventional internal combustion engine/automatic transmission systems	Electric propulsion system	15-year life, capable of delivering at least 55 kW for 18 seconds and 30 kW continuous.	$12/kW peak (system cost)
	Fuel cell power system (including hydrogen storage)	60% peak energy-efficient, durable system, specific power of 325 W/kg, power density of 220 W/L, operating on hydrogen.	$45/kW (by 2010) [c] $30/kW (by 2015) [c]
Enable clean, energy-efficient vehicles operating on clean, hydrocarbon-based fuels powered by either internal combustion power-trains or fuel cells	Internal combustion systems	Peak brake engine efficiency of 45%, meet or exceed emissions standards.	$30/kW
	Fuel cell systems, including a fuel reformer	Peak brake engine efficiency of 45%, meet or exceed emissions standards.	$45/kW (by 2010) [c, d] $30/kW (by 2015) [c, d]
Enable reliable hybrid electric vehicles that are durable and affordable	Electric drive-train energy storage	15-year life at 300 Wh, discharge power of 25 kW for 18 seconds.	$20/kW
Enable the transition to a hydrogen economy, ensure widespread availability of hydrogen fuels, and retain the functional characteristics of current vehicles	Hydrogen refueling with commercial codes and standards and diverse renewable and non-renewable energy sources.	70% energy efficiency well-to-pump.	Cost of energy from hydrogen equivalent to gasoline at market price assumed to be $1.25 per gallon (2001 dollars) [e]
	Hydrogen storage systems	Available capacity of 6 wt % hydrogen, specific energy of 2000 Wh/kg, and energy density of 1100 Wh/L.	$5/kWh [f]
	Internal combustion systems operating on hydrogen	Peak brake engine efficiency of 45%, and meet or exceed emissions standards.	$45/kW by 2010 $30/kW in 2015

Improve the manufacturing base	Material and manufacturing technologies for high-volume production vehicles	Simultaneous attainment 50% reduction in the weight of vehicle structure and subsystems, increased use of recyclable/renewable materials.	Affordable

[a] Technology-specific goals for the DOE's FreedomCAR and Vehicle Technologies Program obtained from the DOE FreedomCAR web-site at http://www.cartech.doe.gov/freedomcar/technical-goals.html.
[b] Cost references are based on calendar year (CY) 2001 dollar values. Where power (kW) targets are specified, those targets are to ensure that technology challenges that would occur in a range of light-duty vehicle types would have to be addressed.
[c] Not including vehicle traction electronics.
[d] Including fuel cell stack subsystem, fuel processor subsystem, and auxiliaries; but not including fuel tank.
[e] Targets for hydrogen dispensed to a vehicle assuming a reforming, compressing, and dispensing system capable of dispensing 150 kg/day (assuming 60,000 SCF/day of natural gas fed for reforming at the retail dispensing station) and servicing a fleet of 300 vehicles per day (assuming 0.5 kg used in each vehicle per day). Targets also based on several thousand stations, and possibly demonstrated on several hundred stations. Technologies may also include chemical hydrides such as sodium borohydride.
[f] Based on lower heating value of hydrogen; allows over a 480 km (300-mile) range.

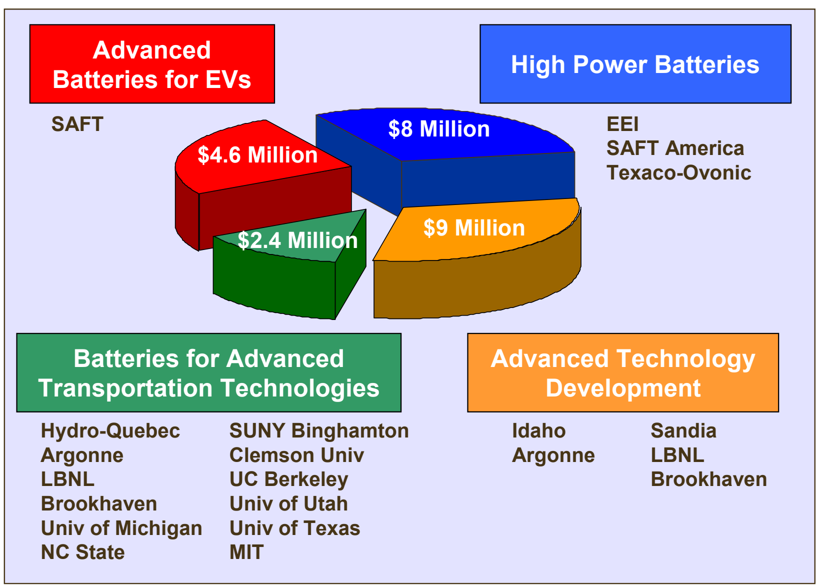

Figure 1: DOE's FY 2002 R&D budgets for energy storage R&D programs

2 Electric vehicle battery research and development program

The goal of the EV Battery Research and Development Program is to support the development of a U.S. domestic advanced battery industry that will meet the USABC technical goals, defined in Table 2. This program has had several major successes, including development and introduction of the nickel metal hydride (NiMH) advanced battery for EV use, resulting in over 1,000 NiMH battery EVs put into service in the past few years. Currently, the program is focused on the development of advanced lithium-based batteries, which is carried out by the U.S. Advanced Battery Consortium (USABC). Specific recent activities are described below.

Table 2: U.S. Advanced Battery Consortium goals for EV batteries

Primary criteria	**Long-term goals[a] (2005 to 2008)**
Power density[b] W/l	460
Specific power[b] W/kg (80% DOD/30 sec)	300
Energy density[b] Wh/l (C/3 discharge rate)	230
Specific energy[b] Wh/kg (C/3 discharge rate)	150
Life (years)	10
Cycle life[b] (cycles)	1000 (80% DOD), 1,600 (@ 50% DOD), 2,670 (@ 30% DOD)
Power and capacity degradation[b] (% of rated spec)	20%
Ultimate price, OEM ($/kWh)(10,000 units @40 kWh)	<$150 (desired to 75)
Operating environment	–30°C to 65°C
Recharge time[b]	< 6 hours
Continuous discharge in 1 hour (no failure)	75% (of rated energy capacity)
Secondary criteria	
Efficiency[b] (C/3 discharge and C/6 charge)[c]	80%
Self-discharge[b]	<20% in 12 days
Maintenance	No maintenance. Qualified personnel service only.
Thermal loss[b]	Covered by self-discharge
Abuse resistance[b]	Tolerant. Minimized by on-board controls
Specified by contractor Packaging constraints, Environmental impact, Safety, Recyclability, Reliability, Overcharge/over-discharge tolerance	

[a] For interim commercialization (reflects USABC revisions of September 1996).
[b] Specific criteria can be found in the USABC EV Battery Test Procedure Manual [1].
[c] Roundtrip charge/discharge efficiency.

2.1 Lithium-ion battery development

The objective of the lithium-ion battery development program is to develop an EV lithium-ion battery system that meets high performance levels for energy and power and has a long life, a low cost, and abuse tolerance. The technology is being developed by SAFT. A summary of the lithium-ion battery system development project appears in SAFT papers [2, 3]. The performance data for this technology was reported in an earlier EVS overview paper [4] and has also been summarized in the EV Battery R&D Program Annual Progress Report [5].

2.2 Benchmark testing of advanced electric vehicle batteries

To conduct a direct comparison of international battery technologies with those developed in the U.S., the Program office has a mechanism in place to conduct testing independently and to hold the results in confidence between DOE and developers/suppliers. Representative NiMH and lithium-ion battery technologies are evaluated using either the procedures in the USABC Battery Test Procedures Manual (for EV Batteries) [1] or in the Partnership for a New Generation of vehicles (PNGV) Battery Test Procedures Manual (for HEV batteries) [6]. During the past fiscal year, batteries representing international technologies were acquired and tested, including Shin-Kobe lithium-ion EV cells (based on lithium-ion technology with manganese dioxide cathodes, rated at 3.75 V and 90 Ah), Panasonic/Matsushita prismatic test pack (based on Ni/MH technology and rated at 144 V and 6.5 Ah), and Shin-Kobe HEV cells (based on lithium-ion technology with manganese dioxide cathode technology and rated at 3.6 V and 3.6 Ah). The tests were conducted at the Electrochemical Analysis and Diagnostics Laboratory (EADL) at Argonne National Laboratory (ANL).

2.3 Workshop on development of advanced battery engineering models

A workshop on the Development of Advanced Battery Engineering Models was held August 14–16, 2001, in Arlington, VA. The purpose of the workshop was to review current research on advanced battery models for HEV and EV applications, emphasizing both applied and basic studies, increase interactions and information exchange between individuals concerned with battery development and packing/applications. The presentations from this workshop are summarized in its Proceedings [7] and detailed papers will be included in a future special issue of the Journal of Power Sources [8].

2.4 Advanced battery readiness ad hoc working group

The Advanced Battery Readiness Ad Hoc Working Group (ABRWG), a forum established to identify, discuss and recommend solutions to barriers in the areas of battery shipping, battery reclamation/recycling, and in-vehicle safety, is composed of governmental officials, private-sector representatives from battery and automotive companies, recycling and chemical-processing companies, and representatives from the electric power partnerships such as the Electric Power Research Institute. During the past fiscal year, the ABRWG held a meeting in Arlington, VA, in February 28 - March 1, 2001. The meeting was devoted to shipping, recycling/reclamation, and in-vehicle safety issues. More detailed information regarding this meeting appears in the EV Battery R&D Program Annual Progress Report [5].

3 Batteries for Advanced Transportation Technologies (BATT)

High cell potentials and demanding cycling requirements for automotive applications of advanced rechargeable batteries lead to important chemical and mechanical instabilities, issues that must be addressed before successfully developing and scaling-up such batteries. Some core DOE development activities on advanced EV batteries with USABC are supported by the Batteries for Advanced Transportation Technologies (BATT) Program, managed by Lawrence Berkeley National Laboratory (LBNL), with the active involvement of other national laboratories, universities, and industrial organizations. It addresses fundamental issues of chemistries and materials that face all lithium battery candidates for DOE EV and HEV applications. The selected battery chemistries are monitored continuously with periodic substitution of more-promising components. The program is organized into six research tasks. Recent BATT accomplishments for each of the above tasks are summarized below. More detailed information on the individual accomplishments appears in BATT progress reports and the corresponding annual report [9, 10].

3.1 Cell development

The cell development task has identified three "baseline" rechargeable lithium cell chemistries. The polymer-electrolyte cell chemistry includes a Li negative electrode, $Li(CF_3SO_2)_2N$ + cross-linked PEO-based electrolyte, and V_6O_{13} or another compatible positive electrode. The gel-electrolyte cell chemistry includes a graphite negative electrode or a high-capacity Sn-based electrode with acceptable stability, $LiBF_4$ + cross-linked gel electrolyte, and a $LiFePO_4$ or $Li_{1.02}Al_{0.25}Mn_{1.75}O_{3.97}S_{0.03}$ positive electrode. The baseline (ATD Program Gen 2) Li-ion chemistry is graphite + PVDF binder negative electrode, $LiPF_6$-EC-EMC electrolyte, and $LiAl_{0.05}Ni_{0.80}Co_{0.15}O_2$ + graphite + acetylene black + PVDF positive electrode. Current efforts aim to develop cells for the testing and characterization of BATT Program baseline and advanced chemistries, including the development of a manufacturing process for electrodes evaluated in test cells, the choice and validation of standardized test cells, development of testing protocols, testing of new components obtained from other BATT Program researchers, and delivery of post-test materials to BATT Program diagnosticians. Another ongoing effort evaluates electrode materials by structural characterization of active components as received (or synthesized), following cell disassembly, and *in situ* during cycling. In collaboration with Hydro-Quebec, the program is also developing low-cost cell materials, in particular modified $LiFePO_4$ cathodes, which exhibit improved utilization and acceptable rate capability. The improved discharge characteristics of $LiFePO_4$ cathodes are shown in Figure 2. An additional project will address the problem of cell venting and evaluate novel approaches to impart overcharge tolerance for BATT Program cells. Recent accomplishments include:

- Lithium-ion pouch cells exhibited a capacity fade rate similar to those of standard-design commercial cells, thereby verifying the viability of the BATT Program pouch cells for the evaluation of advanced cell components.
- An electro-polymerization process was used to prepare a novel switchable current shunt within a porous polypropylene separator, as a means to provide overcharge protection for series-connected Li-ion cells. The shunt was able to carry a current of 5 mA/cm^2 between a stainless steel positive electrode and a lithium negative electrode at a cell potential <5V.

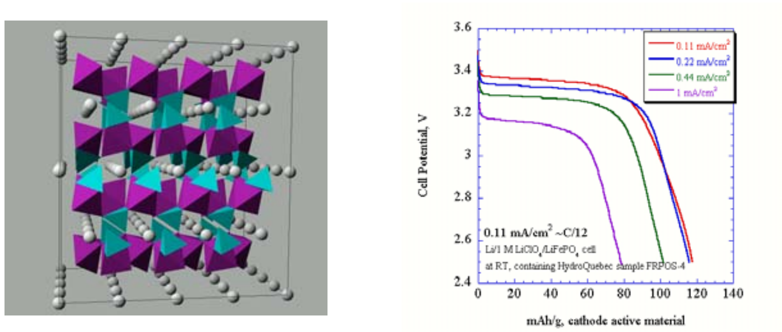

Figure 2: Discharge Characteristics of $LiFePO_4$ Cathodes

3.2 Anodes

The anodes task seeks to characterize and improve graphitic and other carbon materials, as well as conduct exploratory research on non-carbonaceous anode materials. Low-cost metal alloys with acceptable capacity, rate, cycle-ability, and calendar life are currently under investigation. The participants are ANL, State University of New York (SUNY) Binghampton, and University of University of Michigan (UM). Recent accomplishments include:

- Tests of a new prelithiated composite $SnO-LiC_6$ anode showed no first-cycle irreversible capacity loss, as well as a stable capacity greater than 500 mAh/g after 500 cycles at 100% depth of discharge.
- A stable reversible capacity of 300 mAh/g was achieved with a novel MnSb anode, which is being developed as a safer alternative to carbonaceous anodes in Li-ion batteries.

3.3 Electrolytes

Polymer electrolyte research aims to understand performance characteristics by studies of the transport properties of the electrolyte as a function of polymer and salt structure, polymer structural changes as a function of temperature, and interactions at the electrode/electrolyte interface related to transport and chemical/mechanical stability. This task seeks to identify advanced non-flammable electrolytes (NFEs) and electrolyte additives to determine their effectiveness for liquid-based Li-ion batteries. The participants are LBNL, North Carolina State University (NCSU), University of Utah (UU), and Clemson University. Recent accomplishments include:

- A composite polymer electrolyte (CPE) was prepared by adding fumed silica to a low-molecular-weight polymer. A $Li/CPE/V_6O_{13}$ cell was then assembled, and it showed improved charge-discharge cycle performance, electrochemical efficiency, rate capability, and self-discharge characteristics.
- New research projects were initiated at Clemson University (new lithium battery electrolytes based on oligomeric imide salts) and the University of Utah (molecular dynamics simulation studies of polymer electrolytes), as a result of a request for proposals issued in FY 2001.

3.4 Cathodes

The focus of this task is to develop a high-rate and stable MnO_2 cathode. Although Mn is a low-cost constituent, MnO_2 cathodes tend to lose capacity at an unacceptable rate. Current research is directed at understanding the reasons for the capacity fade and developing methods to stabilize this material, as well as evaluating novel forms of MnO_2 cathodes. The participants are ANL, SUNY Binghampton, LBNL, and University of Texas. Recent accomplishments include:

- Reversible ambient-temperature capacities of 140-150 mAh/g were achieved with novel stabilized layered cathodes such as $xLi_2TiO_3 \bullet (1-x)LiMn_{0.5}Ni_{0.5}O_2$.
- Investigations of low-cost LiFePO4 cathodes revealed that the specific method of applying carbon to impart high rate capability is not an important factor in cathode preparation.

3.5 Diagnostics

This task uses post-test analyses and several enhanced techniques to investigate morphology, structure, and compositional changes of electrode materials. Detailed investigations are also underway to provide a better understanding of the solid electrolyte interphase (SEI) layers that form on electrode surfaces and of the lithium/polymer interface. The participants are LBNL, BNL, and the Massachusetts Institute of Technology (MIT). Recent accomplishments include:

- Raman microscopic studies of tested BATT Program anodes not only revealed the presence of disordered carbon in anodes taken from cells that had lost significant capacity, but also showed that a thick solid electrolyte interphase (SEI) layer containing inorganic products was strongly associated with the local regions of disordered carbon.
- The electrochemical oxidation of candidate Li-ion battery solvents and additives was studied using the rotating ring disk electrode method, *in situ* infrared reflection absorption spectroscopy, and density functional theory. The theory and experiments were in complete agreement that CO (and not CO_2) is the energetically favored decomposition product, a result which has important implications for understanding the decomposition mechanisms of battery electrolyte components.

3.6 Modeling

Models are being advanced to elucidate the failure mechanisms of lithium battery components and to understand the mechanisms for thermal runaway. The participants are LBNL, and the University of Michigan (UM). Recent accomplishments include:

- Battery side reactions were modeled to determine their influence on the measurement of electrolyte transport parameters. It was found that the presence of side reactions can lead to significant errors in the determination of electrolyte transference numbers and activity coefficients, a result which has important implications regarding the reliability of prior measurements, and hence battery analysis and design.
- A detailed model based on statistical simulations was used to predict Li-ion battery electrode conductivities, and the predictions agreed with experimental results. The model accounts for differences in particle shape and amount in the anode, and thus can be used to develop electrodes with enhanced conductivity.

4 High power batteries

A lightweight, compact, affordable, high-power energy storage device is one of the critical pacing component technologies for a viable hybrid electric propulsion system. In contrast to the high-energy requirement of EVs, the energy storage device needed for HEVs must have high specific power; that is, the power-to-energy ratio must be greater than 25 W/Wh, as opposed to 2–3 W/Wh for EVs. Two candidate battery chemistries—NiMH and lithium-based technologies are being investigated under the USABC program. Current program participants include SAFT America, Inc., Texaco-Ovonic (TOBS), Electro Energy, Inc. (EEI), and PolyStor. The primary recent accomplishments are listed below:

- EEI developed a new bladder system for applying uniform NiMH pseudo-bipolar cell pressure.
- TOBS discovered a potential plastic for module construction that demonstrates good moldability and weldability, chemical resilience, toughness, and non-flammability. The material is also recyclable, commercially available, and low in cost. TOBS was also successful in the construction of lower cost system assemblies for NiMH batteries, with a total cost based on 1,200,000 models projected to be $800 or less. Part of the cost reduction is a result of reducing the parts count from >100 to 23 parts/module.
- SAFT has significantly improved the projected calendar life, cycle life, and cold cranking ability for their Li-ion technology with the introduction of new active materials and electrolyte [11]. SAFT claims that these batteries are capable of delivering more than 1000 deep discharge cycles and over 300,000 HEV cycles. A full-size HEV Li-Ion battery was developed for the DaimlerChrysler Town & Country Natrium fuel cell concept-hybrid electric vehicle. SAFT reports that this 64 kg battery has capacity of 2.8 kWh and a typical voltage of 346V. It can deliver more than 54 kW under 18 second pulse discharge at room temperature [12]. Table 3 lists cell characteristics and module performance, as reported by SAFT [12].
- PolyStor demonstrated an innovative cell sealing approach for their low-cost flexible packaging design. PolyStor has improved the cycle life of its gel based lithium polymer system from 50,000 to 200,000, and has estimated calendar life to be up to 10-years.

Additional information on the VHPESP for the past fiscal year appears in a Highlights Report [13].

Table 3: SAFT Automotive Lithium-ion cell and battery performance data[a]

Cell Data		Battery Performance	
Type	HE44	Voltage	
Diameter (mm):	54	Typical	346
Length (mm)		Max regen spike	389
Mass (kg)		Min under HEV	259
Max.Op. Voltage (V)	4.0	Min cold crank	192
C/3 Capacity @Max V (Ah)	44	Energy	
Specific Energy @C/3 (Wh/kg)	140	Total kWh	2.8
Energy Density @C/3 (Wh/l)	275	Typically available %	35%
		Max power @room temp (kW) (discharge: 18s @50% SOC, regen: 2s @50% SOC)	55/49
		Capacity @100% SOC (Ah)	7.25
		Impedance @50% SOC, 50A, Ohms	0.29
[a] Data reported by SAFT [12]			

5 Advanced Technology Development (ATD)

Technical barriers to commercialization of lithium-ion batteries remain in the areas of calendar life, abuse tolerance, and cost. To reduce the R&D risk associated with overcoming these barriers, DOE collaborated with the U.S. auto companies, via the Electrochemical Energy Storage Technical Team, to establish a technical support program at DOE's national laboratories. This national laboratory program, entitled the Advanced Technology Development (ATD) Program, assists the industrial developers of lithium-ion batteries in the development of a low-cost, long-life, safe, and high-power energy storage device that meets or exceeds the technical requirements for power-assist and dual-mode hybrid vehicles.

The ATD Program has three major objectives: (1) to develop and validate the practical application of diagnostic tools at the national laboratories to identify factors that limit calendar life and abuse tolerance for high-power lithium ion technology, (2) to assist the developers in the development of practical solutions, and (3) to develop innovative solutions for reducing cell costs. To address those objectives, the program comprises four tasks which are described below. More detailed information regarding the ATD tasks appears in the Annual Progress Report for ATD [14].

5.1 Life

This task focuses on the development, performance characterization, accelerated aging, and diagnostic evaluation of high-power 18650 cells that were built using different cell chemistries. The Generation 1 (Gen 1) and Generation 2 (Gen 2) baseline cell chemistries are listed in Table 4. A few additional cells were built with a Gen 2 variant C chemistry that used a positive electrode material that contains 10% Al dopant, in place of the 5% Al dopant material used in the Gen 2 baseline cells. The Gen 1 and Gen 2 cell chemistries were selected on the basis of extensive screening tests to identify cell chemistries that are capable of meeting the target power requirements. The screening tests also assess thermal reactivity of the materials, via DSC and ARC tests. Once cells are built, they are used to populate an accelerated aging test matrix or a thermal abuse test matrix. The Gen 2 baseline and variant C cells are currently undergoing accelerated aging and thermal abuse testing. The Gen 2 baseline cells exhibit a much slower rate of cell impedance rise than the Gen 1 cells. The participants in this task are ANL, Brookhaven National Laboratory (BNL), Idaho National Engineering and Environmental Laboratory (INEEL), Lawrence Berkeley National Laboratory (LBNL), and Sandia National Laboratories (SNL). Recent accomplishments include:

- Completed the calendar life aging at 55ºC of 15 Gen 2 baseline cells. Correlation of aged cell data indicates a square-root of time dependence of cell impedance rise for the first 28 weeks followed by a linear rise in power fade in the following weeks to 30 % fade.
- Completed the cycle life aging at 45ºC of 15 Gen 2 baseline cells. Correlation of aged cell data indicates a square-root of time dependence of cell impedance rise for the first 28 weeks followed by a linear rise in power fade in the following weeks to 30 % fade.
- Discovered that Variant C cells built with a cathode material rich in Al show a higher power fade but less capacity fade than Gen 2 baseline cells built with less Al doping.
- Discovered through electrochemical impedance spectroscopy that the power fade in the Gen 2 cells is nearly entirely attributed to an increase in impedance at the electrode / electrolyte interfaces.
- Determined that power fade is accelerated by storage at higher states of charge and higher temperatures.
- Developed an Accelerated Life Test protocol that requires three stages of experimentation in order to minimize the cell count, the time of test, and the accuracy of the prediction.
- Developed a phenomenalogically based electrochemical cell model that incorporates surface film impedance effects. This model will be used with the testing and diagnostic results to test and verify different hypotheses of cell failure.

Table 4: The Gen 1 and Gen 2 cell chemistries

Positive Electrode	Gen 1	By weight: 8% PVDF binder, 4% SFG-6 graphite, 4% carbon black, 84% $LiNi_{0.8}Co_{0.2}O_2$
	Gen 2	By weight: 8% PVDF binder, 4% SFG-6 graphite, 4% carbon black, 84% $LiNi_{0.8}Co_{0.15}Al_{0.05}O_2$
Negative Electrode	Gen 1	By weight: 9% PVDF binder, 16% SFG-6 graphite, 75% MCMB-6 graphite
	Gen 2	By weight: 8% PVDF binder, 92% MAG-10 graphite
Electrolyte	Gen 1	1M $LiPF_6$ in EC/DEC (1:1)
	Gen 2	1.2M $LiPF_6$ in EC:EMC (3:7)
Separator	Gen 1	37 µm thick PE Celgard separator
	Gen 2	25 µm thick PE Celgard separator

5.2 Abuse tolerance

This task focuses on the understanding and mitigation of thermal events that result when high power cells are exposed to abuse conditions. As for now, the primary focus is on thermal runaway initiated by a temperature rise in the cell's surroundings. For testing, Gen 2 cells are externally heated and monitored either in a thermally controlled thermal mass or an accelerated rate calorimeter (ARC). The response is videotaped and the vented gasses are analyzed for chemical composition. To provide further understanding of the chemical events that lead to a thermal response, cell components are evaluated individually or in selected combinations in a differential scanning calorimeter (DSC). As the mechanisms of thermal runaway are elucidated, additives are developed to prohibit or reduce those reactions identified as initiating or greatly accelerating thermal destabilization. The goal is to correlate the chemical reactions with the cell response to thoroughly understand the mechanism(s) that control temperature rise in these cell chemistries. The participants in this task are SNL and ANL. Recent accomplishments include:

- Determined that cell vent gases require the presence of air and a spark source for ignition.
- Determined that there are three phases to cell thermal runaway: 1) 50-125ºC, 2) 125-180ºC, and 3) 180ºC and above.
- Determined that during phase 1, the SEI layer that nominally protects the anode material breaks down allowing the EC/$LiPF_6$ to exothermically react at the lithiated carbon anode surface which leads to cell self heating in an adiabatic environment.
- Determined that during phase 2, $LiPF_6$ and EMC react to form CO_2 causing the cell to vent with the entrainment of electrolyte.
- Determined that during phase 3, the cathode/electrolyte react to cause a major exothermic decomposition that this is almost immediately proceeded by an anode / electrolyte decomposition. These nearly simultaneous events lead to rapid cell disassembly.

5.3 Technology transfer

This task focuses on establishing collaborations with industrial firms to study and develop low-cost industrial-scale processes for manufacturing advanced cell materials. A materials screening process is used to identify life limitations and cost barriers are identified using a cell materials cost model. Suppliers provide their latest materials and cost estimates in exchange for information on how the materials fare in an HEV application. With the help of industrial collaborators, several industrial-scale processes for making multi-doped lithium nickel oxide are being evaluated. Several industrial firms have agreed to produce pilot-scale quantities of our advanced electrolyte systems. This task is conducted by ANL. Recent accomplishments include:

- Established rapid materials screening techniques to evaluate anodes, cathodes, binders, and electrolytes.
- Established collaborations with several industrial firms to study the low-cost industrial-scale processing of multi-doped lithium nickel oxide materials.
- Established collaborations with several industrial firms to study the low-cost industrial-scale processing of stable electrolyte systems.
- Evaluated numerous low-cost natural graphite materials and identified several promising candidates that work in 30% PC-based electrolytes.
- Developed a cost model with a major Japanese materials supplier and identified primary material cost barriers: separator, cathode, and negative current collector.
- Determined through the cost model that great savings can be had if high rate spinels become feasible because far less cell capacity is required to meet the FreedomCAR power assist targets.

5.4 Cost

This task uses the information gained during the screening of prior materials and feedback from the diagnostic evaluations to develop advanced cell materials that will simultaneously help extend the calendar life and enhance the inherent safety of high-power cell chemistries, at reduced costs. The internal R&D efforts have focused on the development of multi-doped lithium nickel oxide cathode materials, advanced electrolyte systems (in collaboration with the Army Research Laboratory), and electrolyte additives. This task also targets the development of unique and novel approaches for reducing the costs associated with the packaging of full-size, high-power lithium-ion cells. The approach is to develop a flexible cell packaging technology to replace the metal cans that are currently used for cell containment. The cell packaging needs to severely limit the permeation of moisture into the cell and the permeation of electrolyte out of the cell in order to achieve the life goal. The approach was expanded during the last year, when the target calendar life goal was increased from 10 to 15 years, to consider the use of organoclay technology to limit the permeation rates for water and electrolyte to levels that will permit the achievement of the 15-year calendar life. In order to go from the current metal cell containment to a flexible pouch, a gel technology must be applied. The gel acts as a glue holding the internal cell components together alleviating the need for solid wall cell containment. Gels typically introduce additional complexities of capacity fade and cell assembly. Several gel systems from developers in Japan and the US are being evaluated to identify those that appear to offer the best combination cost reduction in cell assembly and low rates of power fade. The participants in this task are ANL and INEEL. Recent accomplishments include:

- Developed a Co doped $NiMnO_2$ layered material with reduced area specific impedance, thermal reactivity, and cost.
- Developed electrolyte additives that pre-passivate the negative electrode, which reduce gas generation, protect natural graphite from exfoliation in PC-based electrolytes, and reduce the flammability of the electrolyte.
- Identified a new electrolyte salt that is more stable and costs less than $LiPF_6$. Have demonstrated that Mn spinel cathode materials cycle at 60ºC with little capacity fade and no measurable dissolution when the electrolyte contains this new salt.
- Developed a model based on molecular interactions that allows for the accurate prediction of electrolyte transport properties.
- Established collaborative agreements with 4 separate gel manufactures to make high power cells for laboratory investigation.
- Evaluated several 2nd generation laminates and found that the failure of the adhesive between the electrolyte sealant and the aluminum foil support is the critical barrier to shelf life.
- Installed in-house a twin-screw extruder for making nanocomposite laminate pouches. This should accelerate the effort in developing a suitable combination of clay/polymer laminate that can meet the FreedomCAR 15-year life target.

6 Other activities

To facilitate the success of EV and HEV technologies, the U.S. currently cooperates with other countries, including Japan, specifically with the Lithium Battery Energy Storage Research Association (LiBESRA). DOE is a member of the Executive Committee of the International Energy Agency (IEA) and participates in various Annexes of the Implementing Agreement for Hybrid and Electric Vehicle Technologies and Programs. The U.S. is currently participating in IEA Annex I: "Information Exchange" and Annex VII: "Hybrid Vehicles". It is also participating in Annex VIII "Deployment Strategies" which is operated jointly by two Implementing Agreements, the "Advanced Motor Fuel Implementing Agreement (AMF/IA)" and the "Hybrid and Electric Vehicle Implementing Agreement" (HEV/IA). As part of the Annex activities, DOE attends the Executive Committee meetings held in various countries and also provides status updates on other implementing agreements. In addition to monitoring world-wide developmental activities, DOE also keeps abreast of legislative and regulatory mandates that could impact EVs and HEVs.

7 Conclusion

DOE FreedomCAR and Vehicle Technologies R&D programs in the energy storage area are focusing on high-energy batteries for EVs and high-power batteries for HEVs. The successful commercialization of DOE-funded batteries is a testimony to the success achieved by the DOE USABC cooperative program in accelerating the development of mid-term EV battery technology. Currently, the Phase 3 cooperative agreement continues R&D efforts on lithium-based systems. Lithium-ion technology continues to show promise as a mid-term battery candidate. These and future advances in energy storage technologies will be leveraged with the significant progress achieved in other enabling technologies (such as heat engines, fuel cells, lightweight materials, power electronics, and fuels) to achieve the challenging goals of the program.

The office continually examines its R&D programs to enhance their effectiveness, better leverage available technical and financial resources, and promote information exchange among the various elements. The advanced batteries for EVs and HEVs programs will continue to reassess longer-term technologies that promise performance, life, and cost benefits over the nickel metal hydride and lithium-ion technologies as candidates for future development and use in hybrid propulsion systems.

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