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Key challenges and recent progress in batteries, fuel cells, and hydrogen storage for clean energy systems

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Abstract

Reducing or eliminating the dependency on petroleum of transportation systems is a major element of US energy research activities. Batteries are a key enabling technology for the development of clean, fuel-efficient vehicles and are key to making today's hybrid electric vehicles a success. Fuel cells are the key enabling technology for a future hydrogen economy and have the potential to revolutionize the way we power our nations, offering cleaner, more efficient alternatives to today's technology. Additionally fuel cells are significantly more energy efficient than combustion-based power generation technologies. Fuel cells are projected to have energy efficiency twice that of internal combustion engines. However before fuel cells can realize their potential, significant challenges remain. The two most important are cost and durability for both automotive and stationary applications. Recent electrocatalyst developments have shown that Pt alloy catalysts have increased activity and greater durability than Pt catalysts. The durability of conventional fluorocarbon membranes is improving, and hydrocarbon-based membranes have also shown promise of equaling the performance of fluorocarbon membranes at lower cost. Recent announcements have also provided indications that fuel cells can start from freezing conditions without significant deterioration. Hydrogen storage systems for vehicles are inadequate to meet customer driving range expectations (>300 miles or 500 km) without intrusion into vehicle cargo or passenger space. The United States Department of Energy has established three centers of Excellence for hydrogen storage materials development. The centers are focused on complex metal hydrides that can be regenerated onboard a vehicle, chemical hydrides that require off-board reprocessing, and carbon-based storage materials. Recent developments have shown progress toward the 2010 DOE targets. In addition DOE has established an independent storage material testing center to verify storage capacity of promising materials. These developments point to a viable path to achieving the DOE/FreedomCAR cost and performance goals. The transition to hydrogen-powered fuel cell vehicles will occur over the next 10–15 years. In the interim, fossil fuel consumption will be reduced by increased penetration of battery/gasoline hybrid cars.

Keywords: Batteries; Fuel cells; Hydrogen; Hydrides; Lithium; Hybrid vehicles

1. Introduction

In his 2003 State of the Union Address, President George W. Bush launched the Hydrogen Fuel Initiative to ensure a clean environment and the long-term energy security of the United States. Using hydrogen to fuel the economy can reduce dependence on fossil fuels, diversify renewable and sustainable energy sources, and reduce pollution and greenhouse gas emissions. President Bush's vision is summed in the statement that "the first car driven by a child born today could be powered by hydrogen and pollution-free".

In the 2 years since President Bush launched the Hydrogen Fuel Initiative, the US Department of Energy's Energy Efficiency and Renewable Energy, Fossil Energy, Nuclear Energy, and Science Offices have developed a comprehensive integrated research, development, and demonstration (RD&D) plan identifying the key challenges, activities, and milestones which support a transportation fuel cell commercialization decision by industry in 2015. If this decision is positive, then Americans would be able to choose hydrogen fuel cell vehicles by about 2020.

The US DOE approach comprises near term reduction in oil use by the use of hybrid vehicles for improved efficiency and long-term elimination of oil dependency by hydrogen substitution in fuel cell vehicles. The rate of market penetration of the fuel cell vehicle will determine its impact on future US petroleum consumption. Fig. 1 is based on a possible penetration scenario which assumes a market model of past US transportation fuel transition and assumes that the necessary RD&D to overcome the technical and cost barriers is completed by 2015 [1].

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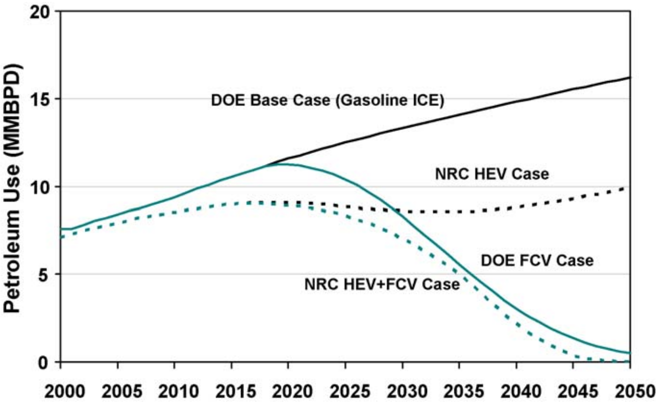

Fig. 1. Vehicle penetration scenarios.

As shown, the gasoline hybrid vehicle will temporarily slow the growth in oil consumption. But as the population continues to grow, gasoline demand will return to historic consumption growth rates. In contrast, the penetration of hydrogen fuel cell vehicles, or a combination of gasoline hybrids and hydrogen fuel cell vehicles, will begin to slow petroleum use and eventually (2020–2025) cause the decline of petroleum use, if a substantial number of light duty fuel cell vehicles are on the road. Note that the projected eventual elimination of oil use in light duty vehicles would not by itself mean that all oil use in the transportation sector would be eliminated, because oil would still be needed for other parts of the transportation system. However, reliance on fossil fuel would be significantly reduced.

Technical challenges for hydrogen fuel cell systems for transportation include cost, durability, and hydrogen storage capacity. Battery systems face challenges in battery cost, performance, life, and tolerance to abuse. This paper will discuss the primary challenges, status, and outlook of development for automotive propulsion batteries, fuel cells, and (on-board) hydrogen storage.

1.1. Advanced energy storage technologies

Batteries are a critical enabling technology for the development of clean, fuel-efficient vehicles. DOE has a major program to develop durable and affordable advanced batteries, in conjunction with its US auto industry partners under the FreedomCAR Partnership, to develop more optimal energy storage devices for use in electric vehicles (EVs), hybrid electric vehicles (HEVs), and fuel cell electric vehicles (FCEVs). Much of this work will transfer to energy storage for heavy hybrid vehicles as well. The large majority of the program focuses on advanced batteries, but the program includes some R&D on ultra capacitors. The advanced battery program is structured as follows:

- Battery Technology Development
    - Full System Development
    - Technology Assessment
    - Benchmark Testing
- Applied Battery Research
- Long-Term Battery Research

Under the Battery Technology Development project, the Full System Development activities are conducted in cooperation with the US Advanced Battery Consortium (USABC). The efforts are focused on developing and evaluating lithium battery technologies and designs for the applications mentioned above. Detailed energy storage targets have been developed for each of these applications, which include several 42 V vehicle systems. The targets for the EV, HEV, and 42 V battery applications are provided in Tables 1–3. Those for FCEVs are still being finalized. This full system development work involves industrial development of advanced battery systems that enable commercially-competitive full-function vehicles for these applications.

In collaboration with the USABC, DOE is supporting the development of high-power Li-ion batteries with CPI/LG Chemical, Johnson Controls Inc., and SAFT, through multi-year contracts with these industrial developers. Additionally, DOE supports the development of low-cost Li-ion battery separators—one of the high-cost items in the battery. These separator multi-year projects are with AMS, Celgard, and UMT. Finally, DOE currently supports the development of ultra capacitors, via a multi-year contract with Maxwell.

Table 1
Energy storage targets for power assist hybrid electric vehicles

Characteristics	Minimum power assist	Maximum power assist
Pulse discharge power (kW)	25 (for 10 s)	40 (for 10 s)
Peak regenerative pulse (kW)	20 (55 Wh pulse)	35 (97 Wh pulse)
Total available energy (kWh)	0.3	0.5
Minimum round trip efficiency (%)	>90 (25 Wh cycle)	>90 (50 Wh cycle)
Cycle life (cycles)	300000 (25 Wh cycle)	300000 (50 Wh cycle)
Cold cranking power at −30 °C (kW)	5 (three 2 s pulses)	7 (three 2 s pulses)
Calendar life (years)	15	15
Maximum mass (kg)	40	60
Maximum volume (L)	32	45
Production price at 100000 year^{-1} ($)	500	800
Maximum operating voltage (V_{dc})	400	400
Minimum operating voltage (V_{dc})	$>0.55 \times V_{max}$	$>0.55 \times V_{max}$
Maximum self discharge (Wh day^{-1})	50	50
Operating temperature (°C)	−30 to +52	−30 to +52
Survival temperature (°C)	−46 to +66	−46 to +66

Table 2
Energy storage targets for 42 V systems

Characteristics	Mild HEV	Power assist HEV
Pulse discharge power (kW)	13 (for 2 s)	18 (for 10 s)
Peak regenerative pulse (kW)	8 (for 2 s)	18 (for 2 s)
Engine-off accessory load (kW)	3 (for 5 min)	3 (for 5 min)
Available energy (Wh at 3 kW)	300	700
Recharge rate (kW)	2.6	4.5
Energy efficiency (% on load profile)	90	90
Cycle life, miles (engine starts)	150000 (450000)	150000 (450000)
Cycle life and efficiency load profile	Partial power assist	Full power assist
Cold cranking power at −30 °C (kW)	8 (21 V minimum)	7 (21 V minimum)
Calendar life (years)	15	15
Maximum mass (kg)	25	35
Maximum volume (L)	20	28
Production price at 100000 year^{-1} ($)	260	360
Maximum operating voltage (V_{dc})	48	48
Maximum open circuit voltage (V_{dc})	48 (after 1 s)	48 (after 1 s)
Minimum operating voltage (V_{dc})	27	27
Maximum self discharge (Wh day^{-1})	<20	<20
Operating temperature (°C)	−30 to +52	−30 to +52
Survival temperature (°C)	−46 to +66	−46 to +66

The Technology Assessment activities are conducted on newly emerging advanced energy storage technologies. These projects with industrial developers are typically 12-month projects or less. They provide the developers with the opportunity to demonstrate the capabilities and potential of their new technologies and provide DOE with the opportunity to assess and validate technical claims of the developers. In some cases a technology assessment project is conducted to determine if a technology is worthy of a full system development project.

The Benchmark Testing activities involve the independent evaluation of advanced cell and battery technologies from around the world. Using resources at its national laboratories, DOE secures and independently evaluates hardware against the manufacturer's specifications and against DOEs energy storage requirements for the most applicable application. Most of these studies are performed under agreements that limit distribution of the evaluation results.

The Applied Battery Research project, denoted the Advanced Technology Development (ATD) Program, is focused on understanding and overcoming the factors that limit the calendar life, abuse tolerance, and operational temperature range of high-power lithium-ion (Li-ion) batteries, while reducing costs at the cell level.

The ATD program utilizes resources available at five DOE laboratories—Argonne, Berkeley, Brookhaven, Idaho, and Sandia—as well as the Army Research Laboratory, to address the four key barriers for high-power Li-ion batteries. The key barriers are: calendar life of 15 years, cost of $20 kW^{-1}, ability to operate between −30 and 52 °C, and to possess sufficient abuse tolerance for use in on-road light-duty vehicle applications. The program focuses on cell-level issues, with the goal of understanding the factors that limit calendar life, abuse tolerance, and performance over the required temperature range. The program attempts to use this material, component, and cell-level knowledge to identify lower-cost materials and components

Table 3
Energy storage targets for electric vehicles

Characteristics	Minimum goal	Long-term goal
Power density (W L^{-1})	460	600
Discharge specific power at 80% DOD (W kg^{-1})	300 (for 30 s)	400 (for 30 s)
Regenerative specific power at 20% DOD (W kg^{-1})	150 (for 30 s)	200 (for 30 s)
Energy density at C/3 discharge (Wh L^{-1})	230	300
Specific energy at C/3 discharge (Wh kg^{-1})	150	200
Specific power/specific energy ratio	2:1	2:1
Total pack size (kWh)	40	40
Calendar life (years)	10	10
Cycle life at 80% DOD (cycles)	1000	1000
Power and capacity EOL degradation (% of BOL)	20	20
Selling price at 25000 units year^{-1} ($ kWh^{-1})	<150	100
Operating temperature (°C)	−40 to +50	−40 to +85
Normal recharge time (h)	6	3–6
High-rate charge (150 W kg^{-1})	20–70% SOC in <30 min	40–80% SOC in 15 min
Continuous discharge in 1 h (% of capacity)	75	75

Table 4
Cell chemistries studied on DOEs ATD program

	Chemistry A	Chemistry B
Anode	MCMB-6/SFG-6	MAG-10
Cathode	$LiNi_{0.8}Co_{0.2}O_2$	$LiNi_{0.8}Co_{0.15}Al_{0.05}O_2$
Electrolyte	1 M $LiPF_6$ in EC:DEC	1.2 M $LiPF_6$ in EC:EMC

Table 5
Organizations participating in the long-term battery research project

Universities	National laboratories	Industrial firms
Brigham Young	Argonne	HydroQuebec (IREQ)
Clemson	Brookhaven	
Michigan	Lawrence Berkeley	
MIT/SUNY at Sony Brook		
North Carolina State/Michigan State		
SUNY at Binghamton		
Texas at Austin		
Utah		

that will enhance the inherent stability of the cell chemistry, thereby increasing the life and inherent safety. Cell-level costs are addressed primarily through the materials and via cell packaging.

To date the program has studied two high-power Li-ion cell chemistries. These cell chemistries are shown in Table 4. They were implemented into high-power 18650 spiral-wound cells, which were subjected to accelerated aging, abuse tests, and post-test diagnostic studies.

The program currently has four focus areas:

- Understand aging mechanisms and more accurately predict life.
- Understand factors that limit low-temperature performance.
- Understand factors that limit inherent abuse tolerance.
- Identify and develop lower-cost and more stable cell-level materials, components, and technologies.

As more information is learned about the factors that limit life, abuse tolerance, and low-temperature performance, this information is used to identify and develop low-cost advanced materials and cell components for simultaneously overcoming these key barriers. The main activities associated with these four focus areas are shown in Fig. 2.

Advanced low-cost cell materials—possessing enhanced chemical, structural, thermal, and electrochemical stability—are being used in a new cell build to study the aging characteristics and inherent abuse tolerance characteristics of another advanced high-power cell chemistry. The positive electrode material is a $Li_{1+x}Ni_{1/3}Co_{1/3}Mn_{1/3}O_2$ material, of the type that is currently beginning to replace $LiCoO_2$ in commercial Li-ion cells for consumer electronic devices in Japan. An improvement in both the aging and inherent abuse tolerance characteristics of high-power Li-ion cells is anticipated. The lithium-rich version of this material, in the form of a composite electrode material, was patented by Argonne National Laboratory.

The Long-Term Battery Research project, denoted the Batteries for Advanced Transportation Technologies (BATT) program, addresses some of the fundamental problems with advanced lithium batteries for transportation applications. The work addresses high-energy batteries for EV applications, as well as high-power batteries for HEV applications. The project involves model development, materials research, and development of diagnostic tools and techniques for studying advanced lithium batteries. The research is performed by universities, national laboratories, and industrial research organizations, as shown in Table 5.

This project addresses fundamental issues of chemistries and materials that face all lithium battery candidates for vehicular applications. It uses several baseline cell chemistries to study their limitations, understand the reasons for these limitations, and use this information as the basis for developing advanced electrode materials and electrolyte systems for use in lithium batteries for advanced vehicle applications. Following are the current areas of activity:

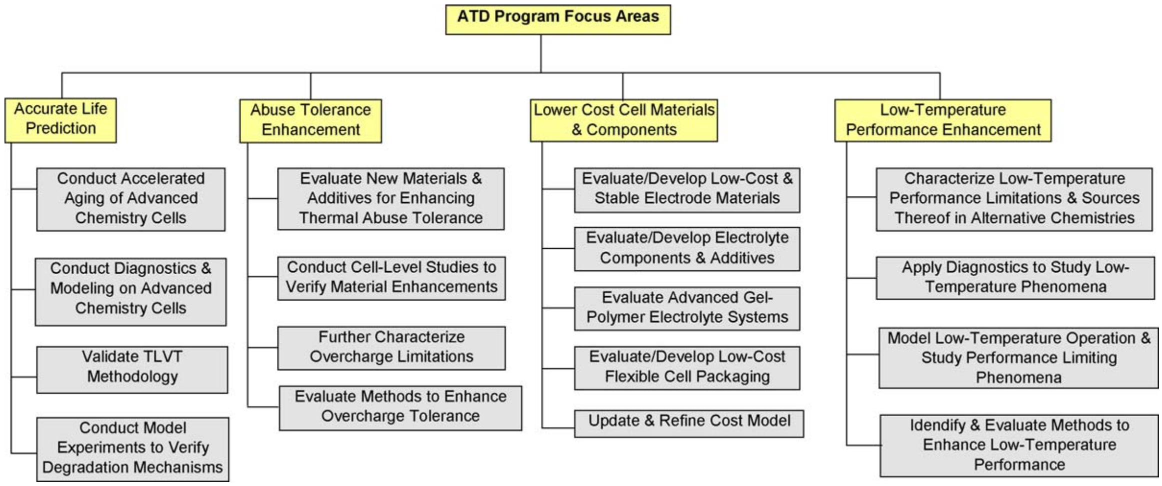

Fig. 2. Main activities in DOEs Applied Battery Research Program.

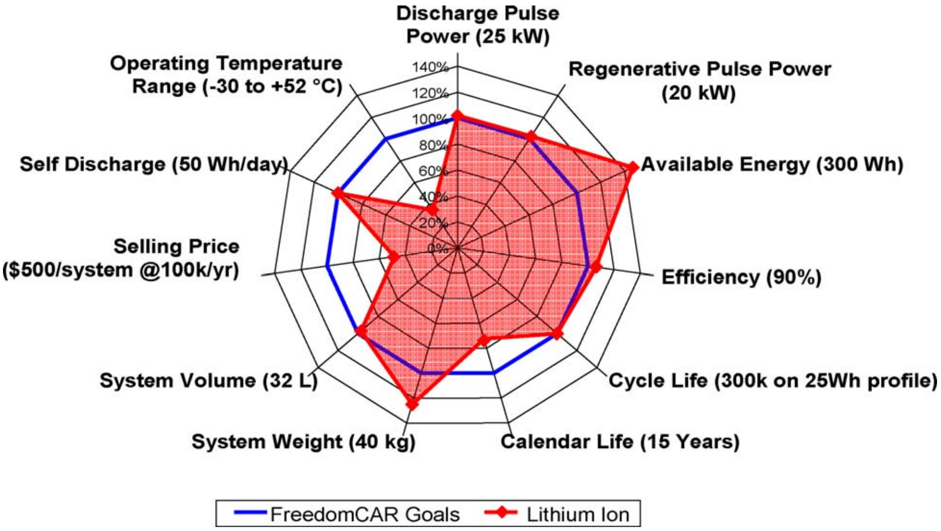

Fig. 3. Spider chart showing status of advanced Li-ion battery system development relative to the FreedomCAR energy storage goals for minimum power-assist HEVs.

- $LiNiCoMnO_2$ and spinel systems—performance and limitations
- Electrolyte limitations
- New high-energy materials
- $LiFePO_4$ system—performance and limitations
- Li/polymer—component limitations

Advanced diagnostic tools and techniques are developed and refined in these studies. These tools and techniques are used to study degradation phenomena in small cells. Results are used as input to empirical and first principle models. Electrochemical cell transport models are developed for each of the baseline cell chemistries and used to study cell performance and degradation processes. Some of the diagnostic tools are used to study the solid electrolyte interphase (SEI) layers (passivation films that form on electrodes during the initial charge half-cycle) and changes in these films during aging. Also, studies are conducted on small cells that employ PEO polymer electrolytes. Molecular dynamic simulations are used to study Li^+ transport through these polymers. Materials research is focused on developing overcharge protection mechanisms, novel cathode materials, and novel anode materials.

2. Battery technology status

While significant progress has been made in developing Li-ion batteries for both HEV and EV applications, major challenges remain. From Fig. 3, calendar life, operating temperature range, and selling price goals for the HEV application are major challenges. Significant progress has been achieved in extending the calendar life and progress continues.

For the electric vehicle application, shown in Fig. 4, calendar life, operating temperature range, and selling price remain challenges. Additional challenges for the EV application include

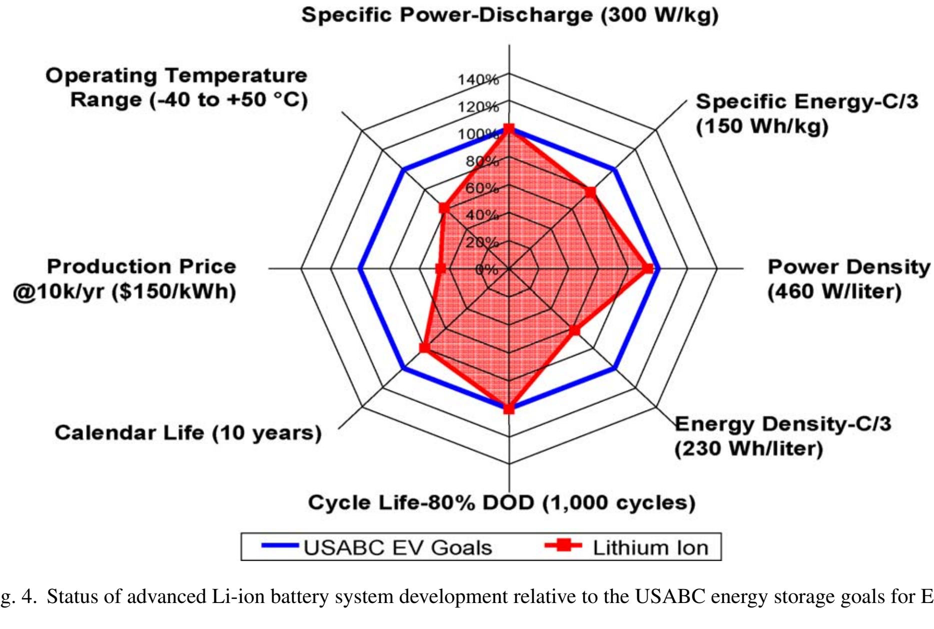

Fig. 4. Status of advanced Li-ion battery system development relative to the USABC energy storage goals for EVs.

Table 6
Transportation fuel cell systems targets and status

Fuel cell transportation systems	Status	2010	2015
System specific power (W kg^{-1})	420	650	650
System power density (W L^{-1})	450	650	650
Durability with cycling (h)	1000	5000	5000
Survivability (°C)	−20	−40	−40
System cost ($ kWe^{-1})	120	35	25

energy density and specific energy. A breakthrough in the development of advanced electrodes (e.g. a higher capacity density positive electrode material) is needed to achieve the energy goals.

2.1. Fuel cells—major challenges

The two most important challenges for fuel cells are cost and durability. The cost for automotive (ICE) power plants is about $25–35 kW^{-1}. Current fuel cell systems are estimated to be about a factor of five higher in cost, even when cost savings for high-volume manufacturing are applied. Major contributors to the cost are the electrocatalyst, the membrane and the bipolar plates.

Automotive fuel cell systems will also be required to be as durable and reliable as current automotive engines, i.e. 5000 h lifespan (150,000 miles equivalent) under heavy load cycling. The performance of current systems decreases substantially after ∼1000 h. While the lifetime for automotive applications is shorter than that for some stationary applications, the cycling requirements make this a more difficult target. The variations in cell potential and relative humidity levels accelerate the degradation of the catalyst layers and membranes. Automotive fuel cells must also be able to function over the full range of vehicle operating conditions (−40° to +40 °C). As can be seen in Table 6, current fuel cell technology does not meet the 2015 targets which would make them competitive with ICE technology.

Fuel cell electrocatalysts are a major cost factor, due to their precious metal content. Estimates of the current cost are at least a factor of 7 greater than the target cost (depending on the cost of Pt assumed). Recent results have indicated substantial progress in reducing the Pt content in the catalysts. UTC fuel cells has decreased the Pt loading by a factor of 2 without a reduction in performance using Pt–Co alloys [2]. These alloys also improve durability, decreasing activity losses and platinum surface area losses in accelerated testing (see Fig. 5).

Other work at 3M has focused on decreasing Pt loading through the use of unique nanostructured thin film (NSTF) catalysts and the use of Pt alloys, and has documented a 5× gain in specific activity over Pt catalysts on conventional high-area carbon supports [3] (see Fig. 6). Using this approach has also increased durability. MEAs made with 3M's NSTF have a lifetime 15–20 times that for MEAs with dispersed Pt/C electrocatalysts.

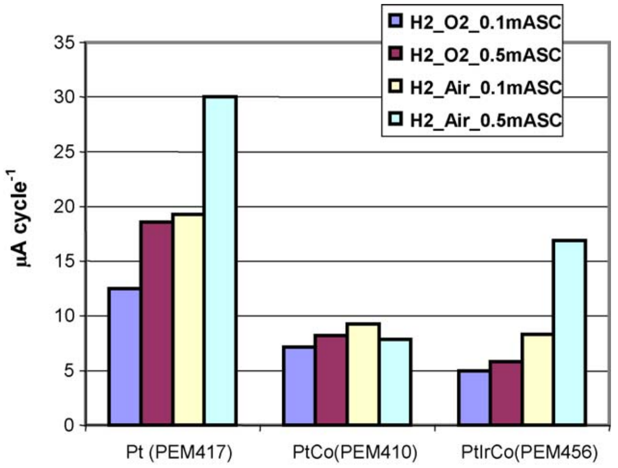

Fig. 5. Performance losses during accelerated testing for MEAs with Pt and Pt alloy catalysts.

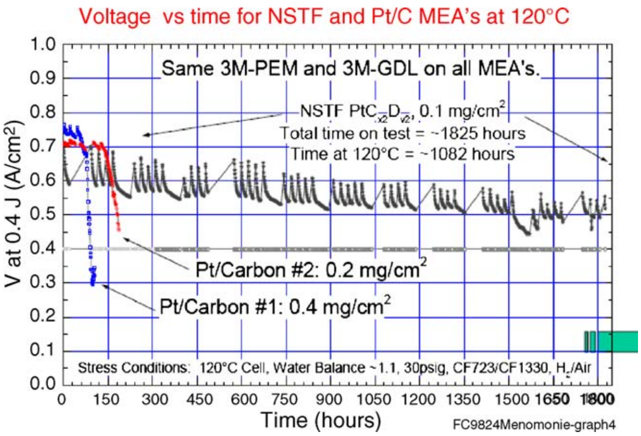

Fig. 6. Accelerated testing of NSTF catalysts showing increased durability.

The polymer electrolyte membranes also offer opportunities and challenges for cost savings and improving durability. Membrane durability is severely challenged by the automotive driving cycle. The combination of potential cycling and variations in humidity of the membrane cause chemical and physical stresses on the membranes and result in tears or pin-holes and membrane failure, well before the 5000 h target lifetime. DOE sponsored work has led to an understanding of the role of peroxide-induced membrane degradation from chemical attack of the polymer end groups, resulting in new polymer membranes with greater durability [4,5]. One example is shown in Fig. 7 [3].

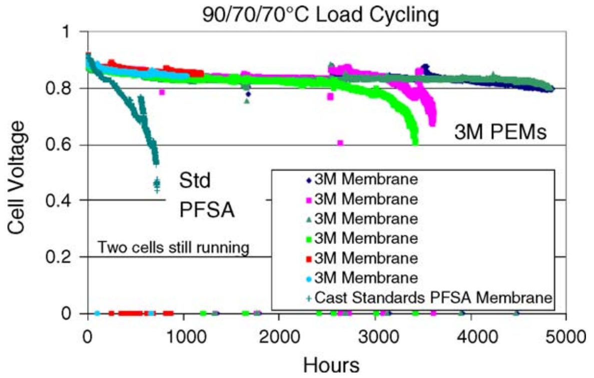

Fig. 7. Increased durability of new 3M ionomers in accelerated testing.

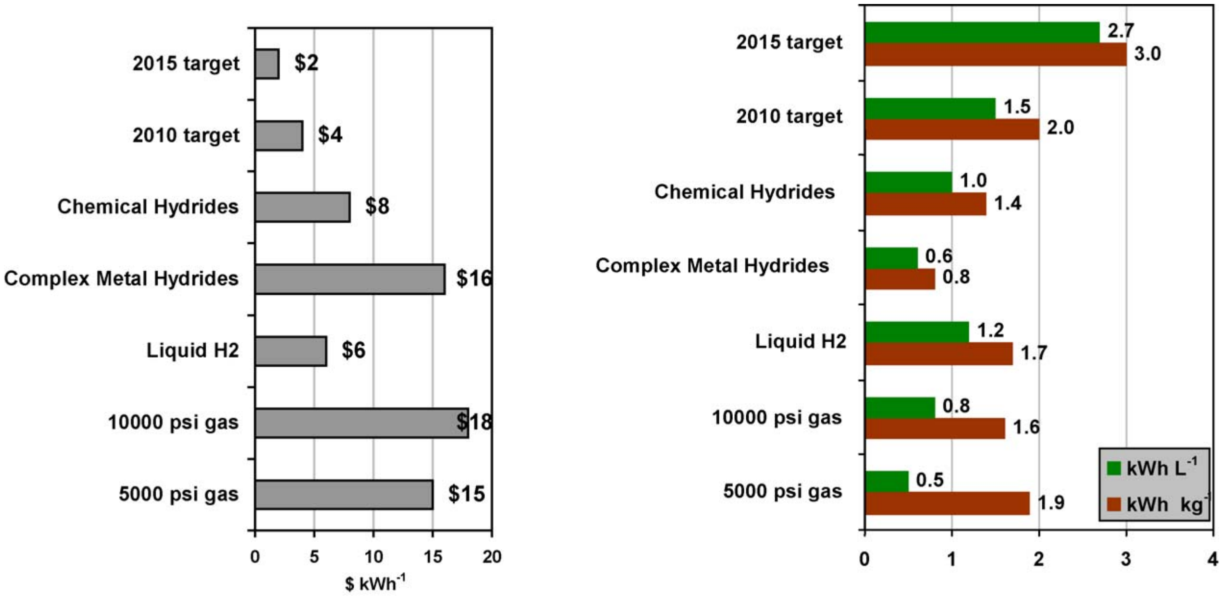

Fig. 8. Status of hydrogen storage systems.

PEM fuel cells must have effective water management systems to operate dependably and efficiently. The ability to operate the membrane at elevated temperatures and lower relative humidity has the potential for reducing costs by simplifying the temperature and humidity control systems. On-going research to develop new membranes, which have high conductivity at low relative humidity and temperatures up to 120 °C has uncovered new membrane systems. However, while these systems have produced membranes with high conductivity under these conditions, other membrane properties such as mechanical strength and chemical stability have not improved to the same degree. New membranes that have all the desired properties are still a challenge.

Another approach to reducing costs is to develop hydrocarbon membranes, which should be less expensive to manufacture than the current state-of-the-art perfluorinated membranes. Recent advances have illustrated some promising systems, such as biphenyl sulfone systems [6] and polyphenylene sulfone copolymers [7]. Again, the proper balance between conductivity, thermal and chemical stability, and physical properties has proven difficult to achieve.

2.2. Hydrogen storage

Hydrogen storage on-board the vehicle is considered key to achieving market success for fuel cell vehicles. To be competitive with ICE vehicles, hydrogen fuel cell vehicles should have a similar driving range. The major challenge is storing enough hydrogen on board for an equivalent driving range of 300 miles while meeting the performance (weight, volume, kinetics, etc.), safety and cost requirements without compromising passenger or cargo space. The energy density is critical. The fuel storage systems in today's vehicles have an energy density about 6 kWh L^{-1}. With the improved fuel economy of a fuel cell vehicle and a conformable hydrogen storage system, the requirement for a fuel cell vehicle is 2.7 kWh L^{-1}. This is a higher energy density than liquid hydrogen (20 K, 1 bar). System studies suggest the hydrogen storage system should also have a specific energy of 3.0 kWh kg^{-1} and a cost of $2 kWh^{-1} to meet the overall goals. No current hydrogen storage technology meets these targets. The status relative to the cost and volumetric and gravimetric energy capacities are shown in Fig. 8.

The storage system adds volume and weight, bringing any systems with current hydrogen storage materials further from the targets. New hydrogen storage materials are needed. The US DOE has initiated the National Hydrogen Storage Project and established centers of Excellence for Metal Hydrides, Chemical Hydrides, and Carbon-based Materials to identify and develop new hydrogen storage materials that can meet the targets. In addition to capacity, the hydrogen storage material must release hydrogen at a relatively low temperature so that the energy needed to release the hydrogen is not a significant drain on the overall system efficiency. The ideal system would be able to utilize the waste heat from the fuel cell to desorb hydrogen from the storage material, but still hold adequate hydrogen at ambient temperatures. The challenge is to tune the materials properties to obtain reversible hydrogen storage systems with properties between the cryogenic hydrogen adsorbents, which have hydrogen bond enthalpies (ΔH) of 4–20 kJ mol^{-1} H_2, and intermetallic and complex metal hydrides which have hydrogen bond enthalpies of 30–55 kJ mol^{-1} H_2.

While no current systems meet the storage requirements, progress is being made. Mg–Li amides have demonstrated a materials-based reversible hydrogen storage capacity of 5 wt.%, with potential for up to 10 wt.%. [8]. Single-walled carbon nanotubes have demonstrated 2.5–3 wt.% hydrogen storage [9]. Theoretical calculations have identified new materials based on the hydrogen bond energies, including cyclopentadiene $ScH_2(H_2)_4$ and PANI conducting polymers [9]. Chemical hydrides such as *N*-ethylcarbazole, have been identified with 5.5–7 wt.% materials based hydrogen storage capacity [10], and amino–borane complexes with mesoporous scaffolds have demonstrated 6 wt.% H_2 capacity, with the scaffold reducing borazine formation [11].

3. Conclusions

The US Department of Energy's Energy Efficiency and Renewable Energy, Fossil Energy, Nuclear Energy, and Science Offices are engaged in a comprehensive integrated research, development, and demonstration program to address the key challenges, activities, and milestones which support a transportation fuel cell commercialization decision by industry in 2015. Significant progress is being made toward meeting the interim (2010) technical and cost objectives of the program and enabling the commercialization and implementation of fuel cell vehicles in the first quarter of this century.

Parallel efforts in the area of advanced batteries will improve cost, efficiency, and durability and enhance the penetration of battery hybrid vehicles. This will result in reduced fossil fuel usage during the transition to fuel cell vehicles operating on hydrogen derived from domestic renewable sources.

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