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Preface

This report contains brief summaries of key accomplishments of the FreedomCAR and Fuel Partnership program for 2004. This program was initiated in 2002 as the FreedomCAR partnership between the United States Department of Energy (DOE) and the United States Council for Automotive Research (USCAR), the latter being a legal partnership comprising DaimlerChrysler Corporation, Ford Motor Company and General Motors Corporation. In his 2003 State of the Union address, President George W. Bush announced a national Hydrogen Initiative. In response to this, the FreedomCAR partnership was expanded in September 2003 to include five energy companies (BP America, ChevronTexaco Corporation, ConocoPhillips, Exxon Mobil Corporation and Shell Hydrogen (US)) and became the FreedomCAR and Fuel Partnership.

The intent of this report is to provide readers with sufficient information to gauge the relevance of each project to the overall FreedomCAR and Fuel initiative goals and the progress being made. These goals are the "Freedoms" originally embraced by the FreedomCAR program:

- Freedom from dependence on imported oil;
- Freedom from pollutant emissions;
- Freedom for Americans to choose the kind of vehicle they want to drive, and to drive where they want, when they want; and
- Freedom to obtain fuel affordably and conveniently.

Previous annual reports, available on the USCAR website at <u>www.uscar.org/freedomcar</u>, summarized some of the significant progress made through the research projects sponsored under the FreedomCAR program. The material was selected from the many hundreds of projects now active and put into the form of single page accounts, arranged by subject matter corresponding to the various vehicle technical teams formed by the partnership:

- Advanced Combustion & Emissions Control
- Electrochemical Energy Storage
- Electrical & Electronics
- Fuel Cells
- Materials
- Vehicle Systems Analysis

Following the addition of the energy companies to the partnership, five additional technical teams were formed:

- Onboard Hydrogen Storage
- Codes & Standards
- Hydrogen Production
- Hydrogen Delivery
- Fuel Pathway Integration

The first two (the "Joint Tech Teams") consist of members drawn jointly from the energy partners, the USCAR partners and DOE, while the last three (the "Fuel Tech Teams") have members drawn from the energy partners and DOE. Progress in projects monitored by the two Joint Tech Teams has been sufficient to justify inclusion of seven accomplishments in this report for 2004. Progress in projects monitored by the three new Fuel Tech Teams will be highlighted in future reports.

In general, each of the 31 accomplishments summarized in this report was selected by the relevant tech team as representing a significant milestone reached, or breakthrough made, in 2004. It represents the fruit of work that may well have begun in previous years. It does not necessarily indicate that the final goals of a particular project have yet been reached.

2004 FreedomCAR Highlight Fast-Throughput Methods Used to Discover New, NOx-Reducing Catalytic Materials

General Motors, Engelhard, Accelrys

Lean combustion conditions favor fuel economy but promote production of nitrogen oxides (NOx), leading to undesirable exhaust gas emissions. There is a need for materials that would be catalytic reduction agents for such NOx.

A promising approach to selective catalytic reduction (SCR) uses hydrocarbons provided by the fuel as the reducing agent. This strategy avoids the limitations of two other approaches: frequent cycling of the stoichiometry of gases in a NOx adsorber; and lack of an infrastructure for urea SCR.

The new method had been rejected previously because the NOx conversion of known materials was too low and reduction of engine-out NOx was too difficult. However, current strategies such as low-temperature combustion have the potential for significant reductions in engine-out NOx. The present work, co-sponsored by DOE, seeks new materials with potentially high NOx conversion.

Such NOx-reducing catalysts consist of combinations of 3-5 materials in varying concentrations. Investigation of the large number of possible combinations of materials and their relative concentrations is not feasible by conventional methods. We therefore used a fast screening process (similar to drug industry practice) to increase the evaluation rate from 1-2 samples per month to hundreds of samples per month. The tests were done on a small sample of a standard honeycomb converter coated with the new material and installed in a flow reactor (the second step in the evaluation after initial screening).

The figure shows the temperature dependence of NOx conversion for several materials, including a new one discovered by fast screening. Its high-temperature conversion is similar to that of the best available NOx catalyst, which is ammonia SCR (ammonia is the result when urea is injected into the exhaust). We expect to improve the low-temperature conversion. The lowest curve shows the best conversion possible from previously known hydrocarbon SCR materials.

It is now feasible to screen and explore many new NOx-reducing catalyst materials that could help to solve the emission problem for lean combustion. The results are being shared with all light- and heavy-vehicle manufacturers.



2004 FreedomCAR Highlight Real-Time Measurement of Particulate Material Sandia National Laboratory (SNL)

Reduction of harmful vehicle exhaust emissions is a major FreedomCAR goal. Tier II emission levels for particulate material (PM) are significantly lower than in previous standards. Although these standards are based on a mass measurement integrated over the test cycle, there is a need for sensitive, realtime PM measurements to guide engine and vehicle development. Since high particulate emissions can occur during transients (e.g., rapid changes in acceleration or deceleration), a fast time response is required. In addition, variation in the composition of the PM, which is affected by engine conditions, requires different after-treatment strategies.

Sandia National Laboratory (SNL) has applied an optical technique, laser-induced incandescence (LII), to measure the elemental carbon portion of PM emissions from a diesel vehicle during transients. The method is not limited to laboratory use and has been commercialized by Artium Technologies.

The figure shows measurements of the PM emissions from a diesel car operated on the road with the system installed in the trunk. The data show the vehicle and engine speeds and the LII signal during an acceleration. There are spikes in the PM during the acceleration and during gearshifts. The system also ran unattended at an engine manufacturer from nearly two months and logged more than 1000 steady-state and transient heavy-duty FPT tests.

Test experiences thus far have demonstrated the robustness and ease-of-use of the instrument. They have also stimulated laboratory development of the system to include measurement of the total PM, along with the elemental carbon fraction, during engine transients. Eventual commercialization of this comprehensive technique is anticipated.



On-Road LII Measurements

2004 FreedomCAR Highlight Response Maps Developed for Lean NOx Traps CLEERS Collaboration

Reduction of harmful exhaust emissions, such as NOx, is a major FreedomCAR goal. The CLEERS (Crosscut Lean Exhaust Emission Reduction Simulation) activity is a voluntary collaboration between light-duty vehicle and heavy-duty engine manufacturers, National Labs, catalyst suppliers, Universities and DOE to improve practices and share pre-competitive information related to lean aftertreatment. Sub-teams focus on NOx adsorbers, selective catalytic reduction of NOx via urea and hydrocarbons, and particulate filtering and oxidation.

One important outcome has been agreement on an industry standard process to understand and represent NOx adsorber performance in a response map that benefits all parties. The supplier of catalyst materials performs a standard, defined set of tests to show catalyst performance; the National Labs, universities, and modelers of catalyst chemistry use such data to refine their models; and the vehicle or engine manufacturer incorporates the data into a simulation for a specific aftertreatment system and predicts its performance.

CLEERS has developed response maps for a NOx adsorber (see below). Each shows the NOx concentration as a function of time in the exhaust of a flow reactor installed in a furnace. In the first part of the test (0 to 15 minutes), the catalyst removes NOx from the lean gases. The NOx concentration at the exit is low initially but increases as the catalyst fills up. At 15 minutes, the stoichiometry of the feed gas switches to rich without NOx to regenerate the catalyst. Depending on the temperature, the trapped NOx is converted to N2 or released without conversion. The response maps show good agreement between the measured and the predicted responses, indicating that the physics of the adsorber is being correctly simulated.

This standardized test procedure and the resulting response maps facilitate investigation of different NOx adsorbers and shortens the development time for aftertreatment systems.



NOx concentration after adsorber for four temperatures (adsorption 0-15 minutes, desorption 15-25 minutes). The red curves indicate the predicted response; the blue curves show the experimental data.

2004 FreedomCAR Highlight X-Ray Measurements Reveal Structure of Diesel Spray Core Argonne National Laboratory (ANL)

A fundamental understanding of fuel combustion in an automotive engine can lead to developments that significantly improve fuel economy and reduce harmful exhaust emissions. Current understanding of the structure of diesel fuel sprays is obtained by optical absorption and scattering measurements with visible light. However, near the injector, the spray density is so high that it completely absorbs the light, making quantitative measurements in this region impossible.

Researchers at the Argonne National Laboratory (ANL) have penetrated the entire region of the fuel spray, not with visible light but with x-rays produced using their Advanced Photon Source facility. These data are the first quantitative measurements of fuel-mass near the spray nozzle as a function of time and position.

The figure shows the evolution of the fuel spray as a function of axial and radial position at several time intervals. The blue areas have low fuel mass and can be studied by visible optical methods. The dark red areas correspond to the largest fuel mass per unit area and are the regions not penetrable by visible light.

Quantitative mass and momentum data for diesel fuel sprays are needed for better understanding of the physics involved and, therefore, improved accuracy of computer models of fuel sprays. These unique near-nozzle data give modelers and injector designers additional insight as to how injector parameters influence the fuel spray structure. The end result should be improved combustion efficiency and lower emissions.



Spray evolution in a 5-bar nitrogen environment with 500-bar injection pressure

2004 FreedomCAR Highlight International Agreement on Hydrogen Fuel Quality Specifications

National Renewable Energy (NREL)

Implementation of a widespread, hydrogen-based, transportation system requires standardization of hydrogen fuel quality. As part of the harmonization of hydrogen and fuel cell standards, the DOE conducted the first international workshop on hydrogen fuel quality for fuel cells in April 2004 and helped the US and Canada establish an agreement with Japan and the European Union on development of hydrogen fuel quality specifications for Proton Exchange Membrane (PEM) fuel cell road vehicles.

This agreement successfully establishes a process that leads to a *temporary* hydrogen fuel quality document as a Technical Specification under ISO. It eliminates another competing effort that would have prematurely established a hydrogen specification that was not temporary. Permanent standards can be addressed in the future, once fuel cell technology is sufficiently developed. The agreement involves amending ISO 14687:1999 (*Hydrogen Fuel Product Specification*) by removing references to PEM fuel cells for road vehicles and adding a Technical Specification for hydrogen fuel quality for PEM fuel cells.

The goal is to have a Technical Specification in place by December 2005. DOE is also coordinating efforts to create a unified international R&D and testing program to obtain the data needed to establish an international standard for hydrogen fuel quality at a later date. DOE and industry are developing a consensus test protocol to obtain the data required to establish hydrogen fuel quality specifications. Single-cell PEM fuel cell testing has now been initiated at the University of Hawaii with test hardware and expertise donated by GM, Ballard, and UTC Fuel Cells



PEM Fuel Cell Test Stand

2004 FreedomCAR Highlight **Predictive Capability Developed for** H₂ Radiation Heat Transfer

Sandia National Laboratory (SNL)

A hydrogen-based transportation system has the potential to achieve key FreedomCAR goals. There is a need to evaluate the potential hazards of using hydrogen and DOE sponsors research to this end, including the development of appropriate national and international standards, codes and regulations based on sound scientific and engineering data.

In FY 2003, experiments were conducted to validate refueling station separation distances with high-pressure releases. A second set of tests at 2000 psi was conducted in FY04. The data obtained were used to validate the models that are employed to quantify hazards. A predictive model for thermal radiation heat transfer based on engineering models and experimentally-verified correlations for large-scale hydrogen jet flames was developed. Based on heat flux limits for protection of people and property, the model is used to define length scales and exclusion zones for various facility designs. An engineering model to determine the hydrogen concentration decay of high momentum, high-pressure, hydrogen jets was also developed. This model can be used to determine the size and footprint of the flammable gas mixture. The figure below illustrates agreement between experimental data and the prediction of the model.

The interim results of the R&D were provided to the International Code Council (ICC) Ad Hoc Committee for Hydrogen Gas to help define refueling station separation distances. Through application of models and parametric studies, Sandia helped the ICC to quantify length scales set by refueling station hazards and develop a separation distance table concept based on working pressure ranges.



Centerline Radiant Heat Flux -- Turbulent Hydrogen Flame

2004 FreedomCAR Highlight Prevention of Premature Codes and Standards Setting Los Alamos National Laboratory (LANL)

Performance-based and harmonized international codes, standards and regulations are critical to the implementation of hydrogen and fuel cell vehicles, at production volumes, with fair and open competition in worldwide markets for hydrogen and fuel cell vehicles.

Teaming with the Department of Transportation, DOE has played a key role in redirecting efforts that could have resulted in premature design-specific regulations for on-board hydrogen storage. By active intervention through the United Nations/Economic Commission for Europe (UN/ECE) World Harmonization of Vehicle Regulations (referred to as WP.29) and its efforts on hydrogen and fuel cell vehicle regulations, further consideration of two draft design-specific regulations on liquid and compressed hydrogen storage was suspended. The US, in collaboration with representatives from Europe (primarily Germany) and Japan, led an effort to develop a roadmap to a global technical regulation (GTR).

This roadmap effort is strongly supported by the leadership of WP.29 as a path forward to a promising hydrogen future. An analysis of the impact on self-certification and type approval processes for each of the four proposed routes is to be conducted by the US (for self-certification) and Germany (for type approval), in an effort to provide balanced information to the WP.29 delegates who will make the ultimate decision on the route to GTRs for hydrogen and fuel cell vehicles.



International Landscape of Vehicle Legal Requirements and Standards (EIHP2)

2004 FreedomCAR Highlight System-on-a-Chip (SoC) Motor Controller Automotive Integrated Electronics Corporation (AIEC)

Hybrid electric vehicles (HEVs) and fuel cell vehicles (FCVs) are of significant interest as ways to address FreedomCAR goals. However, real time control of the permanent magnet electric motor that would be used in such applications is a major challenge.

The powertrain controller in a current internal combustion engines typically contains a moderately powerful microprocessor, which acts as the brain of the control system. It is critical to a network of sensors and actuators, and is the communications link to the vehicle as well as an on-board engine and powertrain diagnostics system. In HEVs or FCVs, it must be complemented or replaced by the controller used for the permanent magnet electric motor that becomes part of the powertrain system or the entire powertrain.

In 2004, AIEC led a group project on the feasibility of designing a motor controller on a single chip that could handle the predicted signal processing, feedback control, communications and diagnostics requirements. The potential reduction in parts count and manufacturing efforts could reduce costs by 40-60% in high volume production.

We have now completed our Phase 1 objectives, a detailed cost study and analysis of potential processes and packaging methods to meet the technical requirements. The design was based on a high-end microprocessor available in the market today.

The study results are encouraging and are under evaluation by the E/E Tech Team members. This Phase 1 design will then be compared to other alternatives before continuation into Phase II, the hardware prototyping of this controller chip.



High-Level Functional Block Diagram of SoC Motor Controller

Analysis of Battery Thermal Performance

National Renewable Energy Laboratory (NREL)

Thermal management of energy storage devices such as batteries is critical for achieving life, performance, cost, and safety target/goals of energy storage for vehicle applications under the FreedomCAR program. However, there has not been a systematic analysis of the factors involved, particularly with advanced batteries, like the lithium-ion (Li-ion) system.

The National Renewable Energy Laboratory (NREL) launched a program in collaboration with battery developers to provide an electro-thermal modeling approach for analyzing the thermal performance of energy storage devices such as batteries.

The resulting finite element approach allows developers to predict the thermal performance of a cell from its electrical behavior. Engineers use their knowledge of cell geometry, material properties, and resistances to calculate the heat generated in the cell and predict temperature distributions based on external boundary conditions and power loads. They can then use this analysis to evaluate the thermal performance of a variety of cell and module designs and technologies.

The electro-thermal model was applied to two Saft lithium-ion cell designs being developed under the FreedomCAR program. Hot spots were identified in the M-HEV cells under transient high-power profiles and led to recommendations that should improve the temperature uniformity and thermal design of future generations of Saft cells.



Analysis of Current Density and Temperature Distribution in Saft Li-Ion Cell

Increased Calendar Life for Li-Ion HEV Battery SAFT USA

Previously, the only Li-ion technology (cobalt-rich nickelate [baseline]) known to have acceptable calendar life had poor thermal stability. Most efforts to improve this stability led to the use of a Mn-rich nickelate material. This showed promise of similar power and energy densities but only poor calendar life (typically 50% of the Co-rich materials).

We wanted to improve the calendar life of the Mn-rich material and develop a positive electrode whose performance equaled that of the traditional Corich materials but which also had enhanced abuse tolerance.

SAFT identified a material with enhanced thermal stability and with initial power and energy densities comparable to those of the baseline material. Evaluation and analysis of samples from independent suppliers revealed that initial material agglomeration determines material cohesion and is a primary factor in subsequent calendar life. SAFT has now incorporated this characteristic into the material specification.

Tests show materials with good agglomeration to have a calendar life of at least 10 years, a major step toward the goal of a 15-year hybrid battery life.



1micron



1micron

Improved material NC5-S4 with good agglomeration and NC5-S3 with nonoptimum agglomeration

Li-Ion HEV Battery Separator Cost Reduction ENTEK Membranes LLC

The cost of components such as the separator in lithium-ion (Li-ion) batteries is currently too high for such batteries to be widely used in hybrid electric vehicles (HEVs).

The objective of this program was to develop a chemically stable, lithiumion battery separator that could be sold for \$1/m² while simultaneously meeting the United States Advanced Battery Consortium (USABC) requirements for electrical, mechanical, and thermal performance.

As part of this program, ENTEK Membranes LLC focused on formulation changes and equipment improvements that enabled it to increase throughput and reduce cost in its continuous manufacturing operation. The samples supplied to SAFT met all performance targets.

Prior to this work, the mass-production price had been estimated to be more than $2.50/m^2$. This assumes an annual volume of greater than five million square meters of a single type of HEV Li-ion separator, sufficient to supply 100,000 vehicles per year.

ENTEK's updated financial model, based on the manufacturing improvements, indicates that its new Teklon[®] separator could be profitably sold for \$1.20/m². This is less than half of the original price and nearly 87% of the way towards the program target of \$1/m².



On-line slitting of 1700 m Teklon[®] rolls onto 3" ID cardboard cores

Ultracapacitor Test Manual

Idaho National Engineering & Environmental Laboratory (INEEL)

Ultracapacitors could play a critical role in Hybrid Electric Vehicles (HEVs). However, there is no standard test protocol for ultracapacitors used in hybrid automotive applications.

The object of this work was to identify appropriate automotive applications, determine key technology hurdles, and develop uniform standards for characterization and testing. A task force was formed to do this and produce appropriate ultracapacitor specifications and test procedures.

As a result of this work, INEEL has published the first Ultracapacitor Test Manual (available on the USCAR website). It defines the ultracapacitors required for FreedomCAR and gives characterization, performance and calendar- and cycle-life tests and analytical methodologies for three core automotive applications: 12V Start-Stop, 42V Start-Stop, and 42V Transient Power Assist. It also includes tests designed specifically to evaluate performance directly against these three core applications as well as more generic tests to evaluate performance over a broader range of interests. The scaling methodology provided enables fair and consistent comparison of the performance of all ultracapacitors regardless of their size, design or specific chemistry.

System Attributes	12V Start-Stop (TSS)		42V Start-Stop (FSS)		42V Transient Power Assist (TPA)	
Discharge Pulse	4.2 kW	2s	6 kW	2s	13 kW	2s
Regenerative Pulse	N/A		N/A		8 kW	2s
Cold Cranking Pulse @ -30°C	4.2 kW	7 V Min.	8 kW	21 V Min.	8 kW	21 V Min.
Available Energy (CP @1kW)	15	Wh	30 Wh		60 Wh	
Recharge Rate (kW)	0.4	4 kW	2.4 kW		2.6 kW	
Cycle Life / Equiv. Road Miles	750k / 150,000 miles 750k / 150,000 m		150,000 miles	750k / 150,000 miles		
Cycle Life and Efficiency Load Profile	UC10		UC10		UC10	
Calendar Life (Yrs)	15		15		15	
Energy Efficiency on Load Profile (%)	90		90		90	
Self Discharge (72hr from Max. V)	<4%		<4%		<4%	
Maximum Operating Voltage (Vdc)	17		48		48	
Minimum Operating Voltage (Vdc)	9		27		27	
Operating Temperature Range (°C)	-30 to +52		-30 to +52		-30 to +52	
Survival Temperature Range (°C)	-46 to +66		-46 to +66		-46 to +66	
Maximum System Weight (kg)	5		10		20	
Maximum System Volume (Liters)	4		8		16	
Selling Price (\$/system @ 100k/yr)	40		80		130	

FreedomCAR Ultracapacitor (End of Life) Requirements

2004 FreedomCAR Highlight Air Compressor Design Meets Performance, Weight, and Volume Targets Honeywell

Internal combustion engines pump their own air, but unusually compact, light, inexpensive, and efficient air machines are needed to supply air to fuel cells to support the oxidation reaction that releases the fuel energy from hydrogen.

DOE convened a panel of experts whose recommendations led to challenging, but realistic, revised targets at the component level (the component targets are subsets of the FreedomCAR goals for the complete fuel cell system). Honeywell responded by designing a new compressor/expander/motor (CEM) unit that is projected to meet the major targets (see below).

The new CEM operates smoothly over a wider range of conditions. It is projected to require less input power than previous machines. Its motor and controller units have significantly reduced weight and volume. Modular design and a reduced parts count have led to reductions in cost.



CEM design projections vs. targets

2004 FreedomCAR Highlight Identification of Fundamental Materials Changes Associated with Fuel Cell Aging

Los Alamos National Laboratory (LANL)

Hydrogen-powered fuel cells are of considerable interest because of their potential role in meeting FreedomCAR goals. However, aside from cost challenges, there are also major technical hurdles that must be cleared, such as fuel cell durability.

LANL launched a project to examine how fuel cell materials change over time in use and how such changes affect performance. Fuel cells were subjected to a series of accelerated tests emulating different aspects of automotive drive cycles to determine which parts of those cycles are most stressful to fuel cells.

The cells were run for extended times both under steady-state operation (constant current) and under the rapidly-changing currents of a standard automotive drive cycle (albeit with the simplification of constant gas flows and humidification). The electrodes were also subjected to repeated cycles of high voltage high potentials (as can occur during shutdown/startup and some types of power transients). The data shown below indicate that this last type of stress has the most dramatic effect on the loss of catalyst active area.

Materials characterization allows correlation of particular failure modes with particular operational stresses. This can guide the development of test protocols to judge the progress of materials and components against the established FreedomCAR system durability targets, thereby indicating pathways to the development of more durable automotive fuel cell systems.

Diverse aging cycles cause different levels of growth of Pt particle size (loss of active area) as detected by X-ray diffraction



2004 FreedomCAR Highlight Low- and No-Platinum Cathode Catalysts Brookhaven National Laboratory (BNL)

Fuel Cell Vehicles (FCVs) offer considerable promise as a way to achieve FreedomCAR goals but the high cost of fuel cells is currently a major barrier to their widespread use in passenger vehicles. A major contributor to the cost is the platinum used in fuel cell cathode catalysts.

Brookhaven's fundamental oxygen reduction kinetic studies have demonstrated possible pathways to replace much or all of the Pt in fuel cell cathode catalysts with other noble metals or noble metal alloys, such as palladium (Pd) or gold-nickel (Au-Ni). Pd and Au have typically been less expensive than Pt and are obtained at least partially from mines in different countries than those for Pt, so the ability to substitute them for Pt could make catalyst supplies more secure.

The laboratory results shown below indicate enhanced oxygen reduction activities for a Pt monolayer $(1\mu g/cm^2 Pt)$ on AuNi particles $(4\mu g/cm^2 Au + 12\mu g/cm^2 Ni)$ on carbon vs. a more standard 10%Pt/carbon catalyst $(12\mu g/cm^2 Pt)$. The Pt monolayer on AuNi particles provides higher activity on a per gram Pt and on a per gram precious metal (Pt+Au) basis, indicating lower cost. Similar Pt monolayer/Pd/C catalysts have been successfully tested in fuel cells out to 2000 hours. These Pt-monolayer materials appear to achieve the activity benefits of Pt-alloy catalysts while having the added advantage that all Pt atoms lie on the active surfaces and are not wasted in the center of the particles.

In preliminary results, BNL has also achieved activities/mass comparable to Pt for a Pd₂Co/C catalyst that completely avoids use of Pt on the cathode.

Comparison of oxygen reduction kinetic currents for Pt₁₀/C catalyst per unit mass of Pt [black], Pt_{ML}/AuNi₁₀/C catalyst per unit mass of Pt [red], and Pt_{ML}/AuNi₁₀/C catalyst per unit mass of precious metal (Pt+Au) [blue]



2004 FreedomCAR Highlight More Durable Membranes for Fuel Cells DuPont / UTCFC/UTRC

Fuel cells powered by hydrogen are expected to play a key role in meeting FreedomCAR goals. However, fuel cells for automotive use operate in a very harsh environment and durability is critical for all of the components, including the membranes in polymer-electrolyte-membrane (PEM) fuel cells.

These thin polymer layers (similar in appearance to plastic food wrap) must be rugged enough to prevent electrical shorting between cells and to prevent pinholes that could mix hydrogen and air. The membranes must also give hydrogen *ions* an easy conduction path from anode to cathode. However, a side reaction in the fuel cell produces hydrogen peroxide and this chemically degrades the membranes over time, although the details of this process are not known. The Department of Energy (DOE) has therefore funded research to identify the processes resulting in chemical degradation.

DuPont not only established that the major degradation is caused by peroxide attack on incompletely fluorinated end groups on the polymer main chains but has also been able to develop membranes that are more resistant to chemical attack. Data (see Figure) taken by collaborator United Technologies Corporation (UTC) show that these chemically stabilized membranes give a decreased decay of the open circuit voltage and substantially improved survival times in accelerated fuel cell tests.

Cooperation between UTC and DuPont has also established the importance of mechanical stresses on membrane life. Stacks with mechanically improved design, using chemically- and mechanically-stabilized membranes, are currently under test in the DOE program. These programs have therefore significantly helped to improve membrane durability and, hence, fuel cell robustness.



Improved Membrane Durability Demonstrated

2004 FreedomCAR Highlight Pt-Alloy Catalysts with Cycling Durability Better than Pure Platinum

UTC Fuel Cells

Fuel cells are expected to play a key role in vehicles that satisfy FreedomCAR goals. However, significant challenges must be overcome, including those of cost and durability. The platinum (Pt) catalyst, critical to the oxygen reduction reaction, is a major contributor to cost and there is therefore considerable interest in reducing the platinum content without sacrificing performance.

It is known that the pure Pt content of these catalysts, by use of Pt alloys, can be reduced by a factor of two without loss of initial oxygen reduction activity. Although this augurs well for potentially significant cost reduction, there could be a durability problem. It has been suggested that corrosion of the non-noble-metal components of these alloys might release ions that would destroy the hydrogen ion conductivity of the membrane and catalyst layers, degrading fuel cell performance over time.

Fundamental work at Lawrence Berkeley National Laboratory demonstrated in 2003 that skin layers of pure platinum could be formed on the surface of cylindrical alloy electrodes. This gave hope that alloy-catalyst corrosion could be avoided. UTCFC has now synthesized practical high-surface-area Pt-alloy/carbon fuel cell catalysts. Laboratory fuel cell tests show (see Figure) that the durability is even better than that of pure Pt, while the oxygen reduction activity is maintained at twice its level.

These results are extremely encouraging and point a way towards durable, costacceptable, automotive fuel cell catalysts.

Highly active Pt-alloy catalysts lose less voltage than pure Pt under potential cycling tests relevant to automotive conditions



Square-wave cycling 0.87 – 1.2V emulates shutdown/startup and local fuel starvation

2004 FreedomCAR Highlight Structure/Performance Relationships in Fresh and Aged Fuel Cells Oak Ridge National Laboratory (ORNL)

Although the hydrogen-powered fuel cell is expected to play a major role in the achievement of FreedomCAR goals, major technical obstacles remain. High power densities are needed and the submicron structure of fuel cell catalyst layers must be tightly controlled to meet cost targets.

As fuel cells age, changes occur in the structure and distribution of the platinized-carbon catalyst, the ionomer (hydrogen-ion-conducting polymer, similar to that used in the membrane) and gas-conducting open pores within the catalyst layers. These changes can have a major impact on durability but research has been hampered by lack of adequate characterization methods.

ORNL has developed transmission electron micrographic (TEM) techniques to fill this need. In collaboration with other national labs and fuel cell developers, we have correlated microstructure with fuel cell performance and durability. The figure shows digitally-processed TEM cross-sectional images of two fresh electrodes prepared by Los Alamos with ionomer contents of 28.6% (left) and 40% (right). The maps show areas of the gas-conducting open pore volume in green, the electron-conducting Pt-coated carbon catalyst in black, and the hydrogen-ion-conducting ionomer in red. The 28.6% sample gave better fuel cell performance. This electrode has more large pores for improved gas access and shows a higher contact area between the ionomer and catalyst phases.

Post-test TEM has also followed migration of catalytic metals during fuel cell operation. Such data clearly show the value of these new techniques for structural characterization.



Phase structure of better (left) and worse (right) performing electrodes

(color code: platinized-carbon, ionomer, open pores)

2004 FreedomCAR Highlight Ternary Pt-alloy Electrocatalysts: Approaching Activity Goal of 4x Platinum Cabot Superior Micropowders

The high cost of the platinum catalysts in current fuel cells is a major barrier to their use in mass-production vehicles designed to meet FreedomCAR goals. The problem could be alleviated if one could find materials that were less expensive than, but had catalytic activity as effective as, pure platinum (Pt).

First-generation Pt-alloy catalysts have already demonstrated twice the oxygen reduction activity of pure Pt. These results are encouraging and show a path toward the 2015 FreedomCAR targets for cost and Pt content. Even higher activities per gram Pt are needed to overcome (1) any shortfalls against the other fuel cell technical targets or (2) higher-than-assumed Pt prices (\$450/troy ounce). DOE has therefore sponsored research to generate more complex, second-generation Pt-alloy catalysts.

Cabot Superior Micropowders (CSMP) has developed a combinatorial method of synthesizing a large library of multicomponent alloy catalysts of varying composition (via spray-pyrolysis). Subcontractor DuPont conducted rapid activity screening of all these materials. CSMP then fabricated the most promising into small fuel cells for testing under rather more realistic conditions.

The kinetic results on H_2/O_2 (see Figure below) are encouraging but need to be proven in hydrogen/*air* cells. The baseline Pt data shown here fall below the state-of-the-art, so further improvements in electrode structure are needed. However, the data from several levels of testing indicate that such materials may give the required activity, about four times that of pure platinum.

PtM₁M₂ shows 60mV gain (~8x activity improvement) vs. pure Pt in single-cell test of oxygen reduction kinetics in H₂/O₂ cell



2004 FreedomCAR Highlight Unique Nanostructured Thin Film (NSTF) Catalyst Demonstrates Improved Durability 3M

The durability of fuel cell components is currently inadequate for widespread automotive use. Two major mechanisms for the degradation of performance over time of typical fuel cell catalysts are (1) conversion of the many very small (~2nm) platinum (Pt) particles into fewer, larger particles with less active surface area and (2) corrosion of the high-surface-area carbon (C) support that initially keeps the Pt particles separated.

3M has now developed a unique electrode structure, consisting of a thin continuous layer of Pt deposited onto whiskers of a nonconductive polymer that has the potential to avoid these problems.

The structure shown below has only about one seventh the Pt surface area per gram of Pt of that of conventional Pt/C catalysts and therefore might be expected to have lower activity. However, kinetic studies both at 3M and at national laboratories show that the Pt thin films of this structure initially have about five times the activity per unit Pt surface area of the small, isolated Pt particles of conventional catalysts. Thus the initial kinetic activity per geometric area of the NSTF fuel cell is comparable to that of a conventional system.

Laboratory experiments have shown the NSTF catalysts to be unusually resistant to corrosion of the carbon support both in gaseous and electrochemical environments. They are readily manufacturable as roll goods. If an improved ability to operate under realistically wet conditions can be demonstrated, NSTFbased membrane-electrode assemblies could prove to be unusually durable and cost-effective alternatives to conventional fuel cell structures.

Unique, and durable, electrode structure



Polymer whiskers coated with a continuous thin layer of platinum

Promising Work in Progress

Hydrogen Storage System Pre-prototype Using NaAlH₄ Complex Hydrides

United Technologies Research Center (UTRC)

Hydrogen-fueled vehicles require sufficient on-board hydrogen storage to allow them to achieve greater than 300-mile driving range. The storage system must be able to transfer heat into and out of the material in desorption mode and adsorption mode respectively. It must also be able to contain the elevated hydrogen pressure used in refueling.

Although complex hydrides seem promising, they present engineering challenges not encountered with better-known classical metal hydrides like LaNi₅. The present work seeks to understand the system-level issues of a complex-hydride-based storage-system for vehicular application. UTRC chose catalyzed sodium alanate (NaAlH₄) as the storage medium for this pre-prototype system.

The figure shows a cut-away view of a 1-kg H_2 system based on NaAlH₄ and designed to reversibly store hydrogen at low pressure for an indefinite amount of time. A carbon fiber composite vessel was used because of the 70-100 atmospheres pressure required for acceptable hydrogen charging kinetics. The system must also minimize weight, volume and cost. The calculated gravimetric and volumetric component system percentages shown are for an improved 5-kg H_2 system based on the current pre-prototype design. The tubing and conductivity-enhancing metal foam provide a 15 minute refueling time with only 1% parasitic loss associated with pumping power.



1-kg hydrogen prototype cutaway and mass and volume distribution for a 5-kg system using 1-kg first generation prototype modules.

This project has revealed how the properties of the metal hydride material such as H_2 capacity and reaction kinetics impact both the weight and volume of a system. It is a starting point for exploring future *in-situ* reversible hydrides that may have even higher gravimetric hydrogen storage density.

2004 FreedomCAR Highlight Hydrogen Storage: Establishment of Standardized Testing Facility

Southwest Research Institute (SwRI)

In order to meet the targets for on-board vehicular hydrogen storage, a "National Hydrogen Storage Project" was established with funding for university, industry and national laboratory projects to investigate and develop new metal hydrides, carbon-based materials, chemical hydrogen storage materials, and novel systems. It is imperative that the most promising materials developed in these various laboratories be independently tested and validated using well-established measurement procedures to determine accurate hydrogen storage capacities.

Due to the absence of adequate standard guidelines, dedicated facilities, or certification programs specifically aimed at testing and assessing the performance, safety and cycle life of emerging hydrogen storage materials and systems, an independent and unbiased evaluation facility was established at SwRI in San Antonio, Texas. Its objective is to establish evaluation protocols and standards for the testing and assessment of emerging materials and systems. Upon thorough validation of the experimental equipment and associated protocols, the testing facility and the technical staff that supports it will be available to any prospective innovator as their definitive testing center.

The facility is focused on hydrogen sorption/desorption measurements of small quantities of storage materials but will also handle full-sized storage systems. The ability to test capacity in different ways is critical to ensuring a valid measurement and the laboratory has a mutually complementary suite of capacity measurement techniques, including a magnetically coupled thermogravimetric analyzer (TGA), a Sievert's apparatus and a thermally programmed desorption apparatus. Both volumetric and gravimetric techniques will be available to characterize the performance of storage systems.

SwRI has now commissioned the lab, set up all support and test equipment, and established draft protocols to characterize the performance of hydrogen storage materials. Upon thorough validation, this facility will allow rapid identification of the storage materials and systems most likely to meet FreedomCAR goals.







Promising Work in Progress **Regenerable Off-Board Chemical Hydrogen Storage**

Air Products and Chemicals Inc.

A key challenge to achieving hydrogen-fueled transportation is the ability to meet vehicle packaging and cost constraints while storing enough hydrogen to power a vehicle for greater than 300 miles before it must be refueled. Storage system energy density targets of at least 1.5kWh/L and 6 wt.% hydrogen by 2010, and 2.7kWh/L and 9 wt.% by 2015, are not achievable with today's technology.

Liquid hydrogen carriers allow conformable vehicular tanks, ease of transport and dispensing, and the ability to refuel at ambient temperatures and pressures. With 'chemical hydrogen storage', a liquid may be used to generate hydrogen onboard a vehicle and the 'spent fuel' may be recovered and regenerated off-board to provide the original starting material.

Air Products and Chemicals, Inc. has recently developed a liquid carrier that can release 5.5 wt.% hydrogen at 197 °C. Preliminary results show the material volumetric energy density to be greater than 1.67 kWh/L (or 50 g hydrogen/liter of liquid). Hydrogen is released by a catalytic dehydrogenation reaction that can take place on-board the vehicle using waste heat from some power plants. Water is not required for the reaction, thereby reducing the system's weight, volume and complexity. The new carrier has undergone several cycles with no significant hydrogen capacity loss, important because the material must be rehydrogenated many times. In addition, there are no heat rejection requirements during refueling, a significant advantage over metal hydrides.

Future efforts will include increasing regeneration efficiency and stability, lowering the operating temperature and increasing storage capacity.



Hydrogen generation over time showing 5.5 wt% capacity

Research & Development of High-Capacity On-Board Reversible Materials:

Low-Temperature Reversible Amides/Imides

Sandia National Laboratory - Livermore

Storing sufficient hydrogen on board a vehicle to provide a driving range of 300 miles is a major challenge. An initial goal is to achieve storage system energy densities of at least 1.5kWh/L and 6 wt.% hydrogen by 2010. Materials that can store hydrogen under pressure and release it when heated, such as the lithium amide/imide (LiNH₂ + LiH \Leftrightarrow Li₂NH + H₂) system, are promising candidates.

Sandia set out to validate the claims about lithium amide/imide and modify it to desorb hydrogen at a lower temperature (less than 200 °C) or, equivalently, a higher pressure (at least 0.1 MPa (~1 atm) of hydrogen). A low temperature allows waste heat (from the engine or fuel cell) to be used to release hydrogen; high pressure can avoid the need for a costly, heavy, compressor on the vehicle.

By partial substitution of Mg for Li, Sandia researchers were able to increase the plateau pressure (where the majority of the hydrogen is released) to about 3 MPa (~30 atm) at 200 °C. The reversible capacity currently demonstrated is 4.5 wt.%. This is less than the theoretical limit, offering hope that continued improvements could meet 2010 DOE targets.

The modified van't Hoff diagram below compares the pressure-temperature equilibrium of several well known solid-phase hydrogen storage materials. The most desirable operating conditions for use in vehicles occur in the red box. The dashed blue line shows the new material. Although other materials also fall in this box, they do not have sufficient storage capacity.



2004 FreedomCAR Highlight Light-Weight Front-End Structure Auto/Steel Partnership (A/SP)

Improving fuel economy is a significant aspect in achieving FreedomCAR goals. Reducing vehicle mass is one way to achieve this, provided that vehicle characteristics such as safety and structural integrity are not compromised. The present work was undertaken to see how front-end structural components made from advanced high strength steels (AHSS) compared to conventional ones in mass and performance.

The front rail and bumper assemblies from an OEM-donated vehicle were replaced by components made from Dual Phase 780 and 980 AHS steels respectively. This entailed design changes in the left and right hand rail assemblies: these included octagonal 3-piece tailor-welded inner and outer elements with symmetric flanges, a tapered front section and inner rail reinforcements, as well as a reinforced tailor-welded rail extension. (A tailor-welded part initially comprises individual blanks having only the properties (e.g., thickness, composition) structurally required for their location. They are then welded together prior to the stamping process).

The AHSS system contains only 12 rather than 28 components and achieves a total mass reduction of 23.7% (the goal was 20%). The donor vehicle, fitted with these new components, was subjected to the NCAP 35mph Rigid Barrier impact test. Post-crash inspection showed proper crushing of the bumper and the front section of the rails. No deformation of the vehicle passenger compartment was seen.

Substitution of AHSS for conventional grades of steel can provide significant mass savings when combined with efficient design. The desired geometries are enabled by the improved ductility (and therefore formability) of AHSS. Its greater strength provides the required crash energy absorption properties. The use of tailored blanks saves weight and allows part consolidation, thereby also improving affordability.

	Original mass (kg)	AHSS (kg)	Mass reduction (%)
LH rail assembly	16.5	12.2	26.1
RH rail assembly	16.1	12.2	24.2
Bumper	39.2	30.4	22.4
Total	71.8	54.8	23.7



(original) baseline rail

(new) redesigned rail

2004 FreedomCAR Highlight Light-Weight Magnesium-Intensive Engine Developments

USAMP: Automotive Metals Division (AMD)

Reducing vehicle weight is an important way to improve fuel economy and help to meet FreedomCAR goals. The change from iron to aluminum (AI) engines has already reduced weight considerably. Replacing aluminum with magnesium (Mg) in engine and transmission components would yield even further weight reductions because the mass density of magnesium is about two-thirds that of aluminum.

However, use of magnesium alloys in powertrain components poses significant challenges. Materials must possess high temperature strength and stiffness, creep- and corrosion-resistance and castability, while meeting cost targets.

In 2003, USAMP's Automotive Metals Division (AMD) determined the technical feasibility of Mg V-block engines by completing a finite element model (FEM) of the engine, including fluid dynamics, heat transfer, structural analysis and performance and durability predictions. AMD found that practical Mg-intensive V-6 engine components could achieve almost the theoretical weight reduction (33%) for Mg.

In 2004, the objective was to select appropriate Mg alloys for each engine component and address any issues such as noise, vibration and harshness (NVH) that might result. The Mg alloy selection process was completed, using the mechanical property database already established for the most promising high temperature candidates. Also, the FEM was used to modify the design of each component so as to optimize the NVH performance and minimize weight.

The model shows that we can both achieve the desired NVH performance and maintain the weight reduction of the original design. In 2005, the engines will be assembled using cast Mg components. They will then be tested to validate the performance (including NVH) and durability predictions of the model. Success with the V-engine will be a world first for Mg and a significant step forward for improved fuel economy.



Acoustic Analysis of Mg-Intensive Engine (overall and Mg vs. Al redesign)

2004 FreedomCAR Highlight Low-Cost Titanium Process Feasibility

Pacific Northwest National Laboratory

The demand for better fuel economy is a major reason to reduce vehicle mass. The direct substitution of titanium alloys for steel may provide one way to do this in a number of automotive applications, including fuel cell and internal combustion powered vehicles. The use of titanium in some individual automotive components has reduced their mass by up to 65%. However, the high cost of extracting titanium from the ore has restricted its use to low-production-volume specialty automobiles.

We recently commissioned Camanoe Associates to do a cost study based on new processes for extracting titanium and have begun evaluation of material produced by such methods.

Unlike the current Kroll Process, the new processes can yield titanium continuously, rather than in batches, from the reaction of sodium with titanium tetrachloride. The product can be alloyed directly in a powder form and used for powder metallurgy (P/M) applications.

The Camanoe study indicated that the titanium-rich feedstock is a major cost item and that lower-cost ore feedstocks, such as ilmenite, should be used. Combining low-cost ore feedstock with the new processes could reduce the cost of titanium powder from nearly \$90/lb to below \$2.00/lb.

Initial evaluation of the material produced by this new approach has been encouraging and the projected cost makes it a serious candidate for lightweighting in high-volume-production vehicles.



2004 FreedomCAR Highlight Lower Cost Carbon Fiber for Automotive Structures

USAMP: Automotive Composites Consortium (ACC)

Reducing vehicle weight is an important way to improve fuel economy and help to meet FreedomCAR goals. Historically, carbon fiber-based polymer composites have been used in high performance military aircraft, spacecraft and in sporting goods due to the combination of their high strength and light weight. Structures made from these materials can be as strong as those made from steel but weigh only 15-30% as much.

Their suitability for automotive structures and proven ability to protect drivers during high speed collisions has led to their being the mandated structural material for Indy-type cars. If carbon fiber composites were to be developed for use in mass-production passenger automobiles, vehicle structure weights could be reduced and fuel economies improved.

However, carbon fiber composites are currently too expensive for use in the general automotive passenger vehicle market. The most significant component of their cost is the carbon fiber itself: this is made by the slow thermal pyrolysis of a relatively expensive precursor. The present research projects explored ways to reduce the cost of carbon fibers by choosing alternate starting materials and new methods to process them.

With DOE funding, Hexcel and the Oak Ridge National Laboratory (ORNL) have demonstrated that both textile-based precursors and lignin (an inexpensive waste by-product from paper making) respectively can be used as starting materials. ORNL has also developed a method to carbonize the precursor that uses a microwave-generated plasma. It is much faster, uses less energy and requires significantly less equipment than conventional methods. This processing method will be coupled with more rapid oxidation and stabilization methods under development.

Use of alternate precursors with alternate production methods should reduce the cost of carbon fiber from more than \$8/lb currently to \$3-4/lb, making carbon fiber composites more affordable for some automotive body structures.



Carbon Precursor from Lignin



Microwave Carbonization Unit

2004 FreedomCAR Highlight Recycling Automotive Materials

Argonne National Laboratory, American Plastics Council, Vehicle Recycling Partnership (USCAR)

Innovative lightweight materials are playing a key role in helping to improve vehicle fuel economy and safety. However, they can also present special challenges to recycling.

We have therefore launched a research program under a collaborative research and development agreement (CRADA) to advance technology for sustainable recycling of current and future automotive materials. Its objectives are: 1) to increase the use of renewable/recyclable materials in automotive applications and 2) to remove any recycle barriers that might otherwise preclude the use of advanced lightweight materials.

Studies have begun on a number of technologies, such as mechanical recycling (recovery of materials for re-use in automotive applications), energy recovery, and the conversion of end-of-life vehicle materials (and shredder residue) to chemicals and fuel.

A large-scale pilot materials separation plant was constructed and started up in 2004 in order to demonstrate the technical and economic feasibility of mechanical recycling of automotive materials.

Other work includes analyses of the recyclability of advanced vehicular designs (e.g. hybrid and fuel cell) in order to identify recycle technology gaps.



Pilot Plant at Argonne to Demonstrate Viability of Mechanical Recycling (End-of-life vehicle shredder residue is first processed in a bulk separation unit: the mixed plastics content is isolated and recovered by the froth flotation unit shown above)

Benchmarking Advanced HEV Technology with 4WD Dynamometer Facility

Argonne National Laboratory

Hybrid electric vehicle (HEV) technology can improve fuel economy and thereby help to meet FreedomCAR goals. Increasing interest in and demand for such vehicles has spurred development and production of a variety of HEVs and their subsystems. This has created the need for careful examination and analysis of benchmarking data, a task for which Argonne's Advanced Powertrain Research Facility (APRF) is uniquely suited.

In 2004, we focused benchmarking on Toyota's new Prius HEV, in order to determine the capabilities of this advanced hybrid. A comprehensive test plan was created to evaluate all aspects of hybrid performance and control. Major subsystems (Engine, Battery, Power Electronics, and Auxiliaries) were monitored and nearly 100 tests were performed using a state-of-the-art 4WD dynamometer with SULEV emissions measurement capabilities.

We also devised a new torque measurement device to solve one of the most difficult aspects of vehicle testing, i.e., determining the torque of the motor during engine operation. Conventional systems require substantial modification to the vehicle and limit the test parameters. The new device fits in the existing space between the engine and the drivetrain, thereby avoiding earlier limitations. A low profile set of strain gages mounted on the vehicle's existing flywheel provides the torque measurements. These are then relayed in real time to the data acquisition system in the vehicle.

The new system allows for on-road testing, provides quick, accurate and simple data collection, is applicable to most vehicles, and results in significant cost savings. Design details were first made public at the 2005 SAE world Congress.





2004 FreedomCAR Highlight Powertrain System Analysis Toolkit (PSAT)

Argonne National Laboratory

Numerous new powertrain systems are being examined for fuel cell and hybrid electric vehicles that can offer reduced dependence on fossil fuels. Time and cost constraints preclude the physical building and testing of the many possible configurations. Instead, the necessary evaluations can be done via simulation tools, provided these are sufficiently powerful and accurate.

Argonne National Lab (ANL) has now made major enhancements to the Powertrain System Analysis Toolkit (PSAT) to provide such a simulation tool. These include additional high-fidelity component models (e.g., fuel cell with GCtool, battery with Penn State or diesel engine with Assanis & Associates), a drag & drop user interface and dual energy storage systems.

With its accurate dynamic component models and realistic control strategy, PSAT has been used directly on the test bench and to control test vehicles through its extension for prototyping known as PSAT-PRO. An automated process has also been developed to analyze test data from the Advanced Powertrain Test Facility (APRF) and validate the models.

PSAT has become the reference tool for the vehicle modeling community because of its powerful features as well as its flexibility and reusability. It can be used to meet the requirements of automotive engineering throughout the development process, from simulation to hardware.

PSAT was recently honored with the 2004 R&D 100 award from R&D magazine: this award recognizes the most technologically significant new products introduced into the market each year.



PSAT-PRO