

VEHICULAR HYDROGEN STORAGE USING LIGHTWEIGHT TANKS (REGENERATIVE FUEL CELL SYSTEMS)

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Abstract

Energy storage systems with extremely high specific energy (>400 Wh/kg) have been designed that use lightweight tankage to contain the gases generated by reversible (unitized) regenerative fuel cells (URFCs). Lawrence Livermore National Laboratory (LLNL) will leverage work for aerospace applications supported by other sponsors (including BMDO, NASA, and USAF) to develop URFC systems for transportation and utility applications.

Lightweight tankage is important for primary fuel cell powered vehicles that use on-board storage of hydrogen. Lightweight pressure vessels with state-of-the-art performance factors were designed, and prototypes are being fabricated to meet the DOE 2000 goals (4000 Wh/kg, 12% hydrogen by weight, 700 Wh/liter, and \$20/kWh in high volume production). These pressure vessels use technologies that are easily adopted by industrial partners. Advanced liners provide permeation barriers for gas storage and are mandrels for composite overwrap.

URFCs are important to the efficient use of hydrogen as a transportation fuel and enabler of renewable energy. H₂/halogen URFCs may be advantageous for stationary applications whereas H₂/O₂ or H₂/air URFCs are advantageous for vehicular applications. URFC research and development is required to improve performance (efficiency), reduce catalyst loading, understand engineering operation, and integrate systems. LLNL has the experimental equipment and advanced URFC membrane electrode assemblies (some with reduced catalyst loading) for evaluating commercial hardware (not funded by DOE in FY1999).

LLNL industrial collaborations include: Conformable H₂ storage development with Thiokol Corp., PEM electrolyzer and URFC developments with Proton Energy Systems, Inc. (Proton), and H₂/halogen energy storage demonstration with Solar Reactor Technologies Group (SRT).

Introduction

The LLNL effort to develop electrochemical energy storage systems occupies a crucial regime in the adoption process for hydrogen technologies – between pure research/conceptual feasibility and near-term demonstrations of commercial systems. This effort leaves as many component innovations as possible to others, and seeks to integrate the best systems from the highest performance, readily procurable components. The integration research and component testing being undertaken has already uncovered many operational and design issues that might hinder the use of breakthrough technologies of interest to the DOE and various aerospace sponsors (NASA, BMDO, USAF, and DARPA). A focus on delivering energy storage to the most weight-sensitive applications (aircraft and spacecraft) ensures that key technologies will be properly implemented and combined to perform in real, upcoming vehicle demonstrations.

The two key technologies that LLNL is aggressively implementing are proton exchange membrane (PEM) -based regenerative fuel cells (RFCs) and high-performance tankage for storing compressed hydrogen and oxygen gases. Advanced PEM technology transforms power, while advanced tankage stores energy. Both sets of components must support each other's specifications to deliver breakthroughs in energy storage. Tankage built from available technologies must be lightweight and must cope with volume penalties, gas permeation, and moisture handling to adequately furnish the breakthrough levels of specific energy that RFC systems offer. Such multidisciplinary specifications have yet to be combined in the form of a commercial product. Were it not for LLNL's role as integrator leading industry, and as technical monitor promoting relevant specifications from within DOE-funded demonstration efforts in industry, these functional combinations of component performances would be years rather than months away. In particular, the DOE-funded activities at Thiokol are delivering vehicle-compatible hydrogen test tanks in support of DOE vehicle demonstrations (Golde 1999). The supervision of and close interaction with this industrial demonstration project is one important example of the real effort DOE is sponsoring at LLNL to convert research into demonstrations.

Another DOE-funded industrial demonstration effort, with Proton as prime contractor, has recently been funded to introduce a PEM-based energy storage system into electrical utility applications (Friedland 1999). Besides monitoring this field demonstration, LLNL has been and will continue to be directly supporting Proton's technology development by testing electrolyzer and URFC cell stacks. In the cases of Proton, Thiokol, and Hamilton Standard (which currently offers the most advanced electrochemical components), LLNL has achieved close cooperation with industrial partners who hold the intellectual property. With these partners, LLNL is jointly developing systems relevant to a wide spectrum of applications, as depicted in Figure 1. These systems include high altitude long endurance (HALE) solar rechargeable aircraft (SRA), zero emission vehicles (ZEVs), hybrid energy storage/propulsion systems for spacecraft, energy storage for remote (off-grid) power sources, and peak shaving for on-grid applications (Carter 1998, de Groot 1997, Kare 1999, McElroy 1998, McElroy 1979, McElroy 1977, Mitlitsky 1999-a,b, Mitlitsky 1998-a,b,c,d,e,f,g, Mitlitsky 1996-a,b,c,d, Mitlitsky 1994, Mitlitsky 1993). Energy storage for HALE SRA was the original application for this set of innovations, and a prototype solar powered aircraft (Pathfinder-Plus) set another altitude record for all propeller-driven aircraft on August 6, 1998, when it flew to 80,285 feet (24.47 km) (NASA 1998).

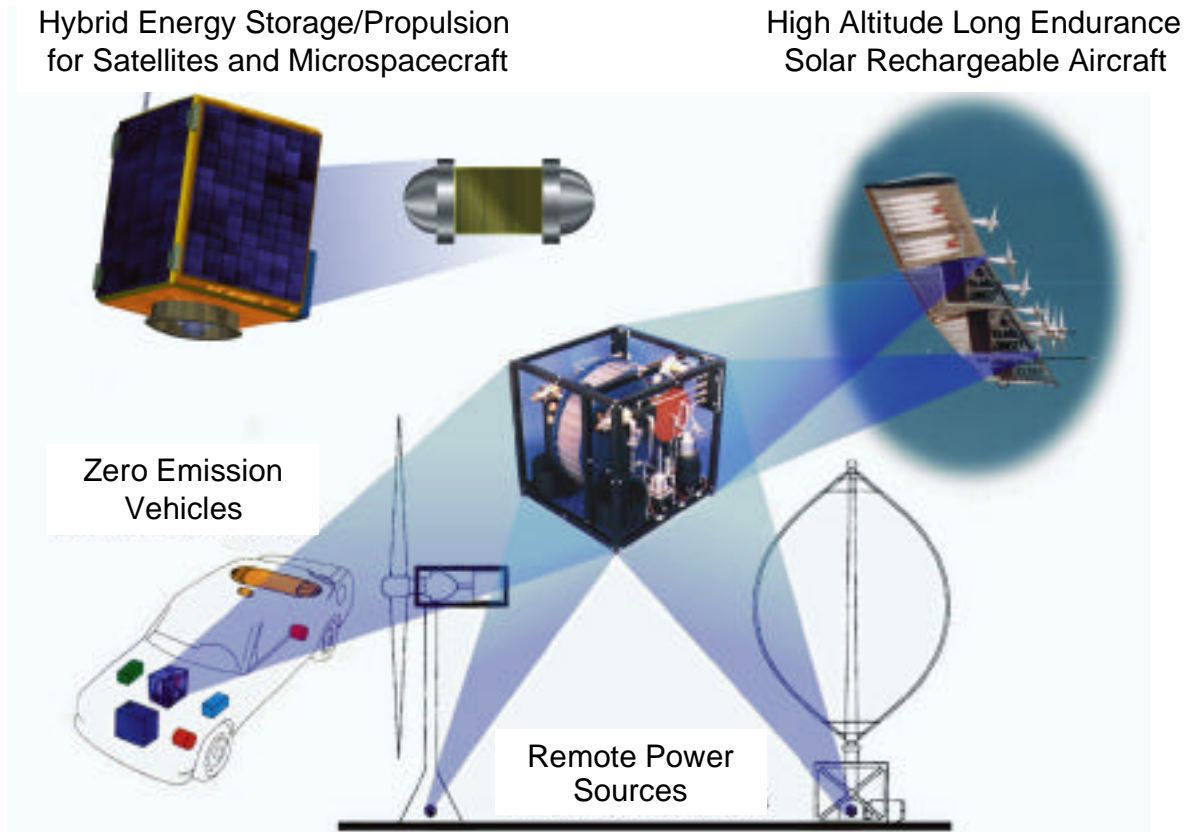


Figure 1 - URFC Systems Have Mobile and Stationary Applications

Hydrogen Storage Goals

Requirements for RFC energy storage systems are similar across a wide range of applications. Although stationary applications generally are not mass sensitive, they join a cluster of mobile, vehicular, and transportable applications that might advantageously store and retrieve energy with RFC systems. Those applications that are most mass sensitive are likely to find RFC systems an enabling technology. All portable systems, all equipment that must ride in land or flight vehicles, as well as the vehicles themselves are in this mass-critical category, as discussed by the LLNL team (Mitlitsky 1999-a, Mitlitsky 1999-b). The LLNL RFC Systems effort is pursuing the most mass sensitive applications where their dramatic specific energy advantages will be clearest compared to secondary batteries. LLNL has also considered RFC systems that use hydrogen/air or hydrogen/halogen chemistries instead of hydrogen/oxygen. Both of these alternatives may emerge as advantageous in some stationary applications, as discussed in the **RFC Systems** section herein.

Mass-sensitivity may be reduced in automobiles compared to spacecraft, but it cannot be ignored. The reason why battery-powered automobiles are not capable of the ~380 mile (610 km) range desired for electric vehicles is due to the mass compounding effect of the energy storage system. Each kg of energy storage on the vehicle results in a 1.3-1.7 kg increase in vehicle mass, due to the additional powerplant and structure required to move and support it. Therefore,

lightweight tankage is required for energy storage systems to have sufficient specific energy in order to achieve the specified range goal. Volume restrictions are an additional constraint on H₂-powered vehicles (RFC, primary fuel cell, or internal combustion) that are not designed from the ground up to accommodate enough H₂ to achieve attractive vehicle range. These vehicle design issues have been studied extensively by Directed Technologies, Inc. (DTI). DTI is in regular and detailed communication with the LLNL team and has provided the technical content of Figure 2 and the spreadsheet in Figure 3 (James 1999). Figure 3 states the assumptions for the 3.6 kg H₂ storage target. Figure 2 shows how H₂ density is related to temperature and pressure and its impact on the DOE 2000 tankage goals. Three overlays of tank external volumes show the relative sizes of tanks (and insulation) which store 3.6 kg of H₂ at 34.5 MPa (300 K), 69 MPa (300 K), and low pressure liquid H₂ (20 K). The non-ideal compressibility of H₂ at high pressures is shown by the decreasing slopes of the density curves (constant temperature) and the sag in the weight % curves (constant tank performance factor). The DOE goal of 12 weight % H₂ at 5,000 psi (34.5 MPa), 300 K translates directly into the need for a tank with performance factor of 1.85 million inch (47.0 km).

DOE 2000 Goals of 12% H₂ by Weight & 700 Wh/liter Relate Directly to Tank Performance Factor, Storage Temperature and Pressure

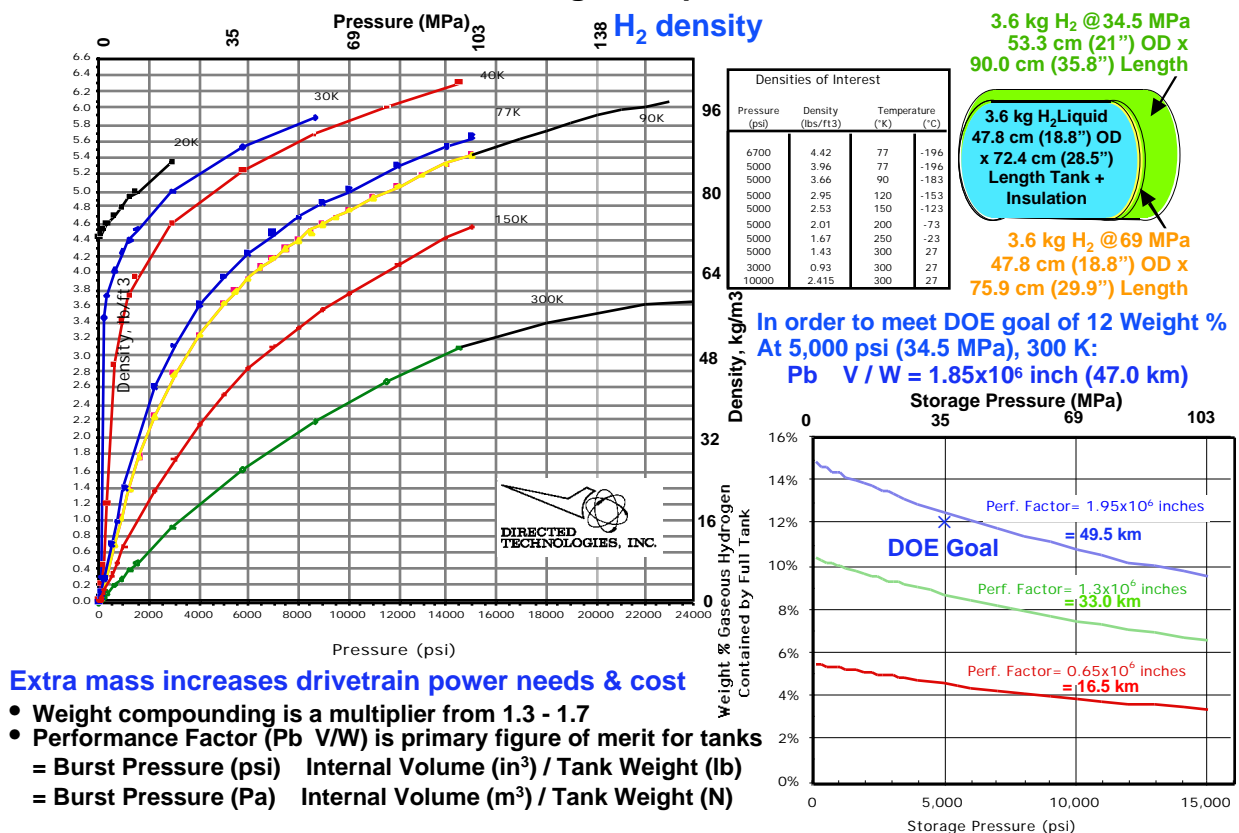


Figure 2 - DOE 2000 Tankage Goals

LLNL has served, and will continue to serve as a conduit for tankage design information between DTI, DOE demonstration programs, and LLNL's industrial partners who are producing high performance H₂ tanks. Besides technical management of the DOE-funded H₂ tankage

development at Thiokol (Golde 1999), LLNL is funded directly by DOE to develop advanced tankage with significantly better performance. Tanks are being built to LLNL specifications, with LLNL design and materials selection which realize the DOE 2000 Goals. No other H₂ storage technology is approaching this accomplishment, particularly in time for its projected feasibility date. Existing H₂ storage mass performance is simply inadequate to provide H₂-fueled vehicles with attractive range, while the DOE 2000 Goal was set to deliver competitive ranges. The spreadsheet in Figure 3 shows the target mass for contained H₂ that DTI provided as input to LLNL's tankage development program. From this table, a stored H₂ mass of 3.6 kg is required for a PNGV-like fuel cell vehicle with a range of 380 miles (610 km) for the EPA Combined Cycle.

		AIV Sable Fuel Cell Vehicle	PNGV-Like Fuel Cell Vehicle	Future Fuel Cell Vehicle
Test Weight (kg)		1344	1032	1032
Drag Coefficient		.33	.27	.20
Frontal Area (m ²)		2.13	2.08	2.00
Rolling Resistance		0.0092	0.0072	0.0072
Fuel Cell Max. Power(kW) (88.5 km/h@7% grade)		39.2	29.8	28.1
EPA	Urban	80.3 mpg	106.2 mpg	112.7 mpg
	Highway	84.8 mpg	113.7 mpg	135.8 mpg
	Combined	82.3 mpg	109.6 mpg	123.1 mpg
1.25xEPA	Urban	69.8 mpg	92.4 mpg	100.5 mpg
	Highway	62.5 mpg	82.4 mpg	102.9 mpg
	Combined	66.5 mpg	87.9 mpg	101.6 mpg
kg H ₂ for 380 miles of EPA Combined Cycle		4.7	3.6	3.1

Figure 3 - H₂ Storage Needs for Battery Augmented Fuel Cell Powered Vehicles

Many of the advanced alternative vehicles this tankage technology enables face an adoption barrier due to the lack of a hydrogen refueling infrastructure. RFC powered alternatives offer the infrastructure independence of recharging from home electricity. Home hydrogen generation by electrolysis also provides a transition to hydrogen fueled vehicles that can precede a hydrogen infrastructure, yet the prospect of buying two PEM subsystems and ensuring their safe operation is likely to be significantly more costly than an RFC system that can reverse chemical to electrical conversion. Such electrically rechargeable vehicles resemble current alternative vehicles powered by secondary batteries, but their weight advantages will give them the range that is currently sacrificed to batteries.

Advantages of URFCs

The energy storage requirements of a solar rechargeable aircraft (SRA) prompted LLNL to commission a study of secondary batteries (Arthur D. Little 1993). Although its predictions for lithium/ion and nickel metal hydride (NiMH_x) batteries have recently been adjusted upward, the basic result still holds strongly in favor of RFC systems. Of these, URFC systems that ‘unitize’ both electrolyzer and fuel cell functions in the same membrane and stack hardware are most mass advantageous. Table 1 compares specific energy, a fundamental performance measure of any energy storage technique, and distinguishes between theoretical and packaged performance for various battery chemistries.

Table 1. URFCs Offer Higher Packaged Specific Energy than Secondary Batteries

Storage System	Theoretical Specific Energy [Wh/kg]	Packaged Specific Energy [Wh/kg]	Comments
H ₂ /O ₂ URFC	3660	400-1000	URFC with lightweight pressure vessels
Li-SPE/MO _x	735	220	Li-solid polymer electrolyte/metal oxide, novel packaging
Ag/Zn	450	200	Excess Zn required for high cycle life, low charge rate
Li/LiCoO ₂	735	150	Poor cycle life, high capacity fade
Li/AlFeS ₂	515	150	400°C thermal management
Na/S	1180	150	~350°C thermal management
Li/TiS ₂	470	130	~50% DOD for high cycle life (900 cycles)
Li/ion	700	100 (135 ^a)	Projection revised Nov. 1996
Ni/Zn	305	90	Excess Zn required, low specific energy
Ni/MH _x	470	70 (85 ^a)	MH _x is metal hydride, projection revised Nov. 1996
Ni/H ₂	470	60	Low specific energy
Ni/Cd	240	60	Low specific energy
Pb/acid	170	50	Low specific energy

^a Projection revised Nov. 1996, private communication, B.M. Barnett (A.D. Little, Inc.)

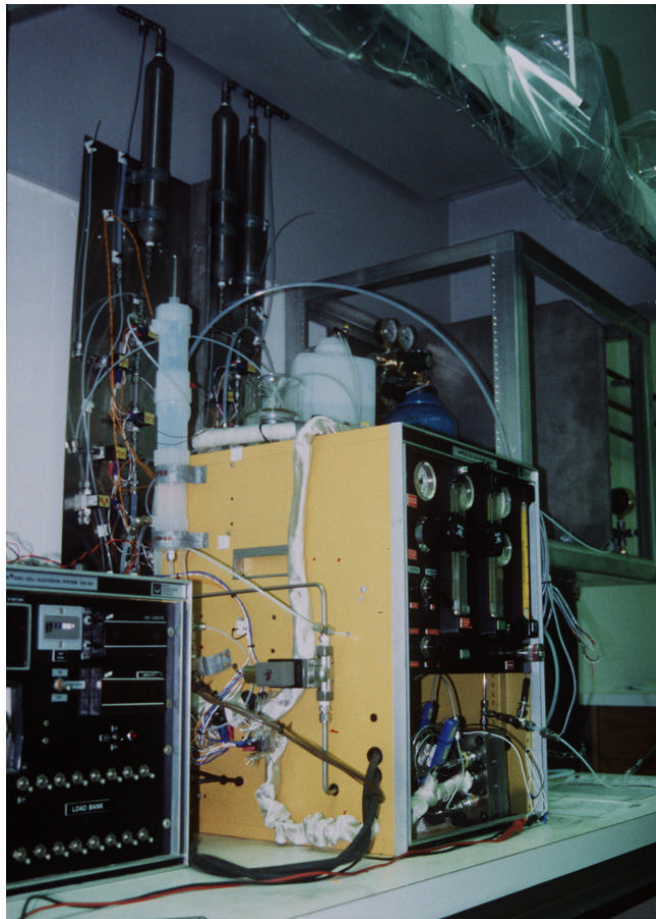
Batteries present a variety of drawbacks that help explain the numbers in Table 1, and provide caveats that make them even less attractive compared to URFC based energy storage systems. Due to reaction kinetics, not all reactants embodied in batteries’ electrodes are accessible, some

cannot be reached and remain as useless weight after discharge. The faster a battery is discharged, the more of its reactants are inaccessible. This effect gives batteries depth of discharge limitations which fuel cells can ignore, since no component in a PEM is chemically changed by the reactions that split water. The absence of consumable electrodes in PEM devices saves them considerable packaging overhead that would otherwise be necessary to support heavy electrodes whose shape changes during charge or discharge. An even more critical advantage of inert electrodes is their avoidance of defect accumulation as they grow or shrink, which not only gives PEM devices long life, it spares them from batteries' trade off between cycle life and depth of discharge.

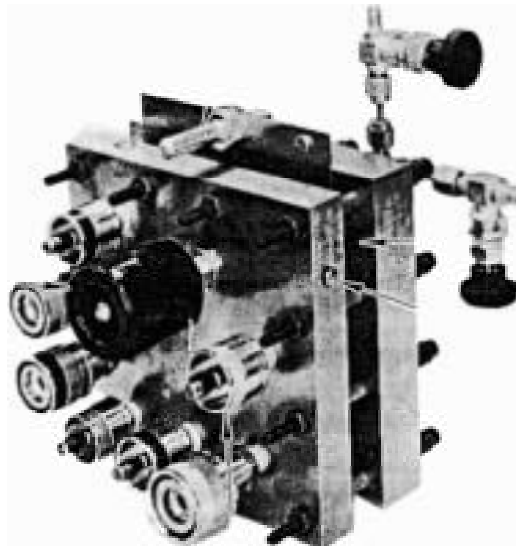
Because fuel cell systems store their reactants outside the electrochemically active cell stack, their specific energy is limited by the mass of the reactant containers. In order to add more energy capacity, these systems must increase their gas and water storage capacity, but the ratings of their PEM stacks need not change. More power capacity is given by simply filling or draining the same containers faster with bigger PEM stacks. Batteries do not offer such independent specification of power and energy, their chemistry forces them to add more heavy reactants in electrodes to increase either rating, burdening their user with unused capacity in the other specification. Reactant containers suitable for PEM RFC energy storage systems do not need to satisfy the complex requirements of battery packaging, but they must be sufficiently chemically inert and impermeable.

LLNL is developing containers suitable for storing gaseous hydrogen and oxygen in the most mass sensitive applications. These containers are pressure vessels derived from aerospace pressure vessel technology. The earliest example of actual, hydrogen impermeable tankage suitable for vehicular energy storage applications is likely to be produced by Thiokol later this year, acting in close collaboration with LLNL under a DOE-funded program. Thiokol is supplying a new tankage technology called 'conformable' tanks, which sacrifice mass performance compared to cylinders but should be ideal for retrofitting the gas tank volumes of existing vehicle designs. Thiokol's configuration has been adapted from designs for compressed natural gas (CNG), that have been reconfigured for hydrogen service. This line of development will be discussed extensively by Thiokol directly in another article in this volume (Golde 1999), so that only its relevance to tankage development directly funded by the DOE at LLNL will be discussed below.

Much of the complexity of battery technology is dispensed with in RFC systems, which don't have to store energetic chemicals as part of the electrode package. LLNL has chosen to explore the URFC, since numerous detailed vehicle mass projections showed energy storage system mass will be significantly reduced by using the same cell stack to convert energy in both directions. Slight efficiency compromises could accompany the choice of a particular set of catalysts (on oxygen and hydrogen sides of a PEM cell membrane) that must serve both electrolysis and fuel cell operating modes. To date, no such performance compromises have been observed, while characterization of unidirectional cell stacks, especially pure electrolyzers, has begun at LLNL in parallel with URFC testing. Figure 4 shows LLNL's existing URFC Test Rig, and its single cell PEM stack, which has been used in previous years to establish the advantages of URFCs. No activity related to URFC development was funded by the DOE in FY1999.



LLNL's URFC Test Rig



URFC Cell Stack (46 cm², 1.1 MPa)

Figure 4 - URFC Test Rig and URFC Cell Stack at LLNL

Although much of LLNL's URFC research has been reported elsewhere (Mitlitsky 1998-b, Mitlitsky 1998-e, Mitlitsky 1998-g), and its lack of FY1999 DOE funding argues against its presentation here, the interests of readers unfamiliar with this technology will be served by the following highlight. URFC technology development continues at LLNL with industrial partners (Mitlitsky 1999-a, Mitlitsky 1999-b), and DOE is supporting PEM electrolyzer work that may lead to a URFC energy storage field demonstration at LLNL's industrial partner, Proton (Friedland 1999). The LLNL team used the apparatus in Figure 4 to refute assertions that URFCs would not be capable of high cycle life, high performance, or reduced catalyst loading. This was done with a series of sensitive measurements that showed essentially no degradation at various current densities across more than 2000 cycles (Mitlitsky 1998-b, Mitlitsky 1998-e, Mitlitsky 1998-g). LLNL has been able to persuade Hamilton Standard (an industrial partner that supports LLNL's aerospace-related energy storage projects) to supply URFC membrane and electrode assemblies (MEAs) with reduced catalyst loading. This constitutes a major success in probing the performance of the best URFC catalysts in aerospace service at economical precious metal levels where this supplier has no current business interest. That research partnership provided the MEAs that were used to measure the electrolyzer (cathode-feed electrolysis) and fuel cell polarization curves shown in Figure 5, which are comparable to the best

(nonregenerative) fuel cells and electrolyzers (Mitlitsky 1998-b, Mitlitsky 1998-e). Thus LLNL has demonstrated the high cycle life, potential low cost, and lack of performance compromises of URFCs.

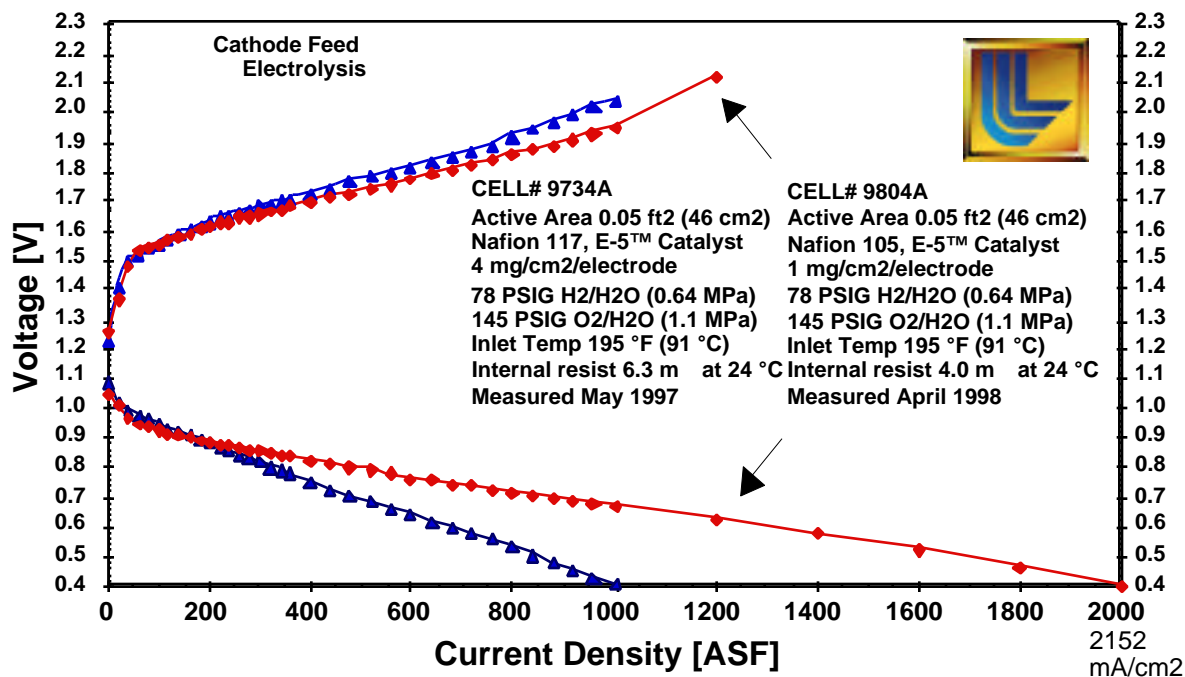


Figure 5 - High Performance URFC Demonstrated With Reduced Catalyst Loading

USAF Funded Electrolyzers

Although not required by any DOE reporting obligations, the interests of readers interested in URFC based energy storage systems will be served by a brief recap of LLNL work performed under USAF sponsorship during the past fiscal year. This work has considerable overlap with portable and solar-powered applications, due to the nature of the small spacecraft which are its intended platform. The electrolyzer considered for this application includes an easy upgrade path to the URFC, and although advanced tankage was developed only slightly during this project, very high tank performance and synergies with vehicle structure will be necessary if later, spaceworthy versions are to be advantageous compared to available space hardware.

A trade study was performed by Hamilton Standard in support of LLNL development of advanced spacecraft for USAF applications. Its focus was electrolysis propulsion for spacecraft (known as a “Water Rocket”), and its deliverables included three preliminary designs of lightweight, high pressure PEM electrolyzers. This small spacecraft propulsion application requires an electrolyzer that would be suitable for charging almost all portable, hydrogen-fueled or oxygen-constrained applications. Static Feed PEM electrolyzers with nominal electrical power inputs of 50, 100, and 200 W, were designed. Table 2 shows that these advanced designs were sized in sufficient detail to predict all component masses. Prior technology that could address the same applications includes vapor feed and high pressure anode feed electrolyzers. In 1986 a vapor feed electrolyzer was assembled with polysulfone frames and a series of electrical and

thermal conducting rings. More recently, anode feed electrolyzers have been tested that provide higher pressure capability than the polysulfone frame design at lower weight per cell. These two types of proven electrolyzers were incorporated into a cell design that supplies high pressure oxygen and hydrogen free of liquid water. This has been accomplished using the Hamilton Standard static feed electrolysis cell configuration and high pressure hardware design.

Table 2. Three Point Designs for Lightweight Static Feed Electrolyzers

Input Power	50 Watt	100 Watt	200 Watt
Number of Cells	16	16	16
Active Area/cell [in ² (cm ²)]	1.39 (8.97)	2.78 (17.9)	5.56 (35.9)
Mass of stack [lb (kg)]	1.15 (0.523)	1.74 (0.789)	2.83 (1.28)
Oxygen pressure [psi (MPa)]	2000 (13.8)	2000 (13.8)	2000 (13.8)
Hydrogen pressure [psi (MPa)]	1980 (13.7)	1980 (13.7)	1980 (13.7)
Water pressure [psia (MPa)]	15 (0.10)	15 (0.10)	15 (0.10)
Operating Temp. [°F (°C)]	70-160 (21-71)	70-160 (21-71)	70-160 (21-71)
Net electrolysis at 120°F (49°C)	2.8 g H ₂ O/hr	5.6 g H ₂ O/hr	11.1 g H ₂ O/hr

Requirements for LLNL's cell stack design included production of hydrogen and oxygen at 2000 psi (13.8 MPa), with water supplied at ambient pressure, long operating life, and as lightweight as practical. Three separate cells were designed for three different power ratings, with active areas of 22.2 in² (143 cm²), 44.5 in² (287 cm²), and 88.9 in² (574 cm²). The 50 W, 100 W, and 200 W units utilize the same 16 cell arrangement, so that the voltage drop for each stack was held constant across all three designs. Each cell is equipped with thermal management provisions in the form of heat conduction tabs to carry heat out of the stack, and power tabs for electrical communication between individual cells, and with the power source. Individual cells are electrically isolated within the stack and connected externally in series. Stack weight in the 100 W unit was minimized with a combination of 16 cells each with a 0.019 ft² (17.7 cm²) active area. For this configuration, the cell outer diameter is 2.63 inches (6.68 cm) and end dome flange diameter is 3.57 inches (9.07 cm).

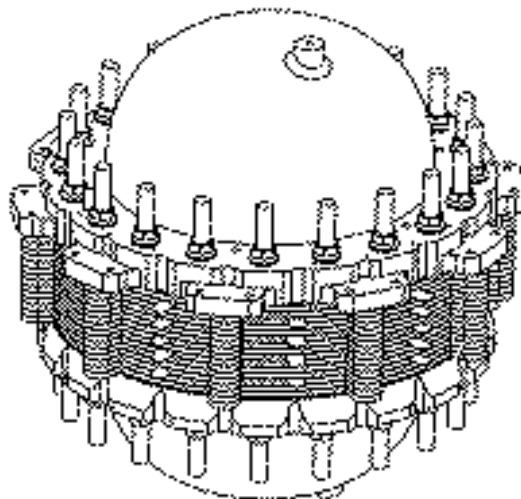


Figure 6 - LLNL Design for 1.3 kg Static Feed 200-Watt Electrolyzer

The 200 W cell stack design is shown in Figure 6. Heat transfer sheets are composed of thin metal foils. Individual cells have a cavity which is ported to the end dome to maintain uniform compression and contact throughout the stack. This cavity removes the need for separate pressure pads, which helps reduce the weight of the cell stack. Stainless steel tie rods and lock nuts are preloaded to compress the seal area and contain the high pressures in the oxygen and hydrogen compartments. The three different sized units use 8, 12, and 20 tie rods respectively. Options to convert this design to a URFC were discussed, although they cannot be presented in similar detail, and amount to roughly a 25% weight to make this design reversible at the same electrolyzer power rating. The low weight of this technology illustrates just how attractive the best aerospace PEM technology could be for portable and mass-sensitive applications. Its existence also argues strongly that high pressure, compact electrolytic generation of gaseous hydrogen from water is a feasible option for refueling hydrogen powered vehicles.

Tankage

The energy-specifying components in LLNL's storage systems have received the lion's share of DOE attention because tanks that meet the DOE 2000 goals are enabling for almost all hydrogen powered vehicles (not just regenerative or fuel cell powered cars). Tankage mass puts a ceiling on stored energy density. Even if the PEM cell stack and ancillaries weigh nothing, system specific energy will be diluted by the mass required to contain reactants. Because cryogenic storage, especially of hydrogen, is such a challenging frontier in itself, a different DOE funded effort underway at LLNL is exploring its expected characteristics (Aceves 1999), and appears documented elsewhere in these proceedings. The significant amount of thermal energy that must be transferred to employ cryogenic hydrogen suggests that cryogenic energy storage media will first find application in primary power plants. The LLNL team pursuing energy storage does not have the energetic luxury to add and recover this significant thermal energy (hydrogen's latent heat of vaporization), so its system integration effort is pursuing the most mass effective storage of gaseous hydrogen and oxygen.

The need for advanced development of compressed hydrogen tankage technology becomes apparent when the specifications commensurate with energy storage applications are combined. Aerospace tankage technology can take advantage of the best composite materials to contain the gas pressure, but relies on liners for permeation control that are massive, ill-conceived, and pose additional mass penalties for cycle life. Without extreme caution concerning the management of residual moisture, the available metal liner options can poison RFC systems. Other forms of tankage simply do not compete strongly with the best composites, as shown in Figure 7. That figure compares a performance factor that is proportional to contained gas mass over tank mass independent of tank scale. (In ideal gases the performance factor sizes tank mass independent of operating pressure as well.) Detailed point designs showed the advantages of developing a tank liner technology that could contain the most reactant (esp. hydrogen) for a given tank mass, and the advantages of a thin liner technology appear in the rightmost bar of Figure 7.

Lightweight H₂ Tanks with Thin Plastic Liners Were Designed to Meet the DOE 2000 Goals Using Easily Adopted Manufacturing Processes

All data (except Carbon/ Thin Plastic Liner and Conformable Carbon / Plastic Liner) from "Onboard Compressed Hydrogen Storage," B.D. James, C.E. Thomas, and F.D. Lomax, Jr., Directed Technologies, Inc., presented at the 1999 Canadian Hydrogen Association Meeting, February (1999)
 Carbon/ Thin Plastic Liner data calculated by Fred Mitlitsky, LLNL, May (1999)
 Conformable Carbon/ Plastic Liner calculated by Richard K. Kunz, Thiokol Corp., May (1999)

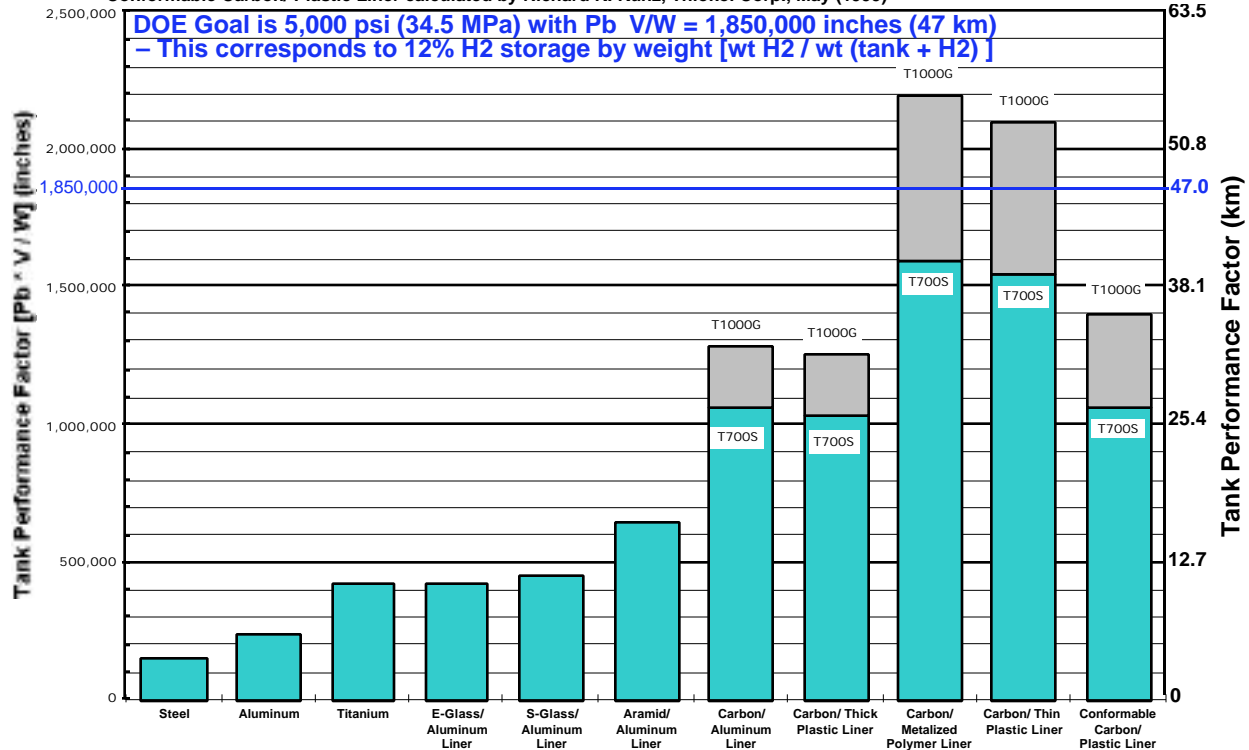


Figure 7 - LLNL Tank Designs Achieve the Required Tank Performance Factor

In order to package gaseous hydrogen into an automobile without enormous changes in vehicle layout, a different approach to tankage makes sense for volume-constrained systems. The LLNL effort is employing DOE funds in a high leverage opportunity to innovate this regime. The tankage technology closest to delivering hydrogen pressure vessels suitable for many mobile applications, including hydrogen-powered passenger vehicles, should soon exist through a development program underway at Thiokol. Thiokol won a DOE contract to develop commercial hydrogen pressure vessel technology for vehicular storage. LLNL serves as technical manager for this development effort, and has intervened repeatedly to insure that Thiokol's developments are most likely to satisfy the true requirements of a hydrogen economy.

Much of the close collaboration between LLNL and the DOE tankage contractors, including Aero Tec Laboratories (ATL) and Thiokol, anticipates the difficult qualification of a liner suitable for resisting hydrogen permeation, matching the process requirements of subsequent composite overwrap, and capable of a high-cycle-life interface with the tank's boss/end detail. Although Thiokol is contributing most of the design, and considerable intellectual property to the boss, all three organizations have learned the necessity of close communication on liner design. Besides the more mass-efficient cylindrical geometry, LLNL's DOE funded development of advanced tanks that will meet DOE 2000 Goals also makes use of Thiokol proprietary boss designs and the rest

of Thiokol's composite winding capability (in order to assure rapid adoption). But the liner technology LLNL is developing must effectively stop hydrogen permeation with a much thinner layer in order to deliver the significant mass advantages forecast in Figure 7. The difficulties of liner material selection were initially underestimated by all the available experts. Many of the relevant material properties the LLNL/Thiokol/ATL team has learned to seek are summarized in Table 3. An experimental program that extends this teams' initial screening process is currently underway which has selected three liner materials suitable to proceed into rotational molding (now underway with new LLNL-owned tooling). This experimental program relies on an expert testing contractor, Southern Research Institute (SRI), that has a long history of furnishing results of Thiokol's specified tests.

Table 3. Literature Survey of Some Polymers Considered for Tank Liners

Figure of Merit = $1 / (\text{Gas Permeability} * \text{Density})$, normalized with HDPE 1

Data are from Modern Plastics 1991 (Mid-October 1990 Issue Vol 67 (11), pp592-6 for Film & Sheet (supplemented with pp480-528 for Resins & Compounds) unless noted
 Permeability to gases data (property #15) is given in units of [cc - mil / 100 in² - 24 hr - atm] @ 25 °C (multiply by 3.886 to convert to [cc - 100 micron / m² - 24 hr - bar])
 Figure of Merit (FOM) is defined as $1 / (\text{Gas Permeability} * \text{Density})$ and is normalized to HDPE (or MDPE if no HDPE data)

Boldface if H2 FOM is >5 (5 times better than MDPE) or if unknown and O2 FOM >5 (5 times better than HDPE)

Boldface and Red if H2 FOM is >25 (25 times better than MDPE) or if unknown and O2 FOM >25 (25 times better than HDPE)

Material	Lo H2 Perm	Hi H2 Perm	Lo O2 Perm	Hi O2 Perm	Lo Density g/cc	Hi Density g/cc	H2 FOM (MDPE=1) 1 / (Lo H2 Perm * Lo Density)	O2 FOM (HDPE=1) 1 / (Lo O2 Perm * Lo Density)	Lo Usable Temp °C [1]	Hi Usable Temp °C [1]	Lo Melt Temp °C [1]	Hi Melt Temp °C [1]
ABS (acronitrile butadiene-styrene)			50	70	1.04		—	3.35				
Acrylonitrile methyl acrylate copolymer, rubberized	240		0.5	0.8	1.15		6.54	302.76			135	
Cellulose acetate	835		117	150	1.28	1.31	1.69	1.16			230	
Cellulose triacetate			150		1.28	1.31	—	0.91				
Cellulose acetate butyrate			600	1000	1.19	1.23	—	0.24			140	
Ethyl cellulose			2000		1.15		—	0.08			135	
Regenerated cellulose (Cellophane)	1.2	2.2	0.5	0.8	1.4	1.5	1074.82	248.69				
ECTFE (ethylene chlorotrifluoroethylene copolymer) e.g. Halar [4]			25	36	1.66	1.7	—	4.19	-160	150	236	246
ETFE (ethylene tetrafluoroethylene copolymer) e.g. Tefzel [4]			100	184	1.7	1.75	—	1.02	-190	150	265	278
FEP (fluorinated ethylene propylene copolymer) e.g. Teflon FEP [4]	2200		715	750	2.15		0.38	0.11	-190	200	260	290
PFA (perfluoroalkoxy) e.g. Teflon PFA [3,4]			296	1142	2.15		—	0.27	-200	250	300	310
PCTFE (polychlorotrifluoroethylene copolymer) e.g. Aclar	220	330	7	15	2.08	2.15	3.95	11.96	-200	180	183	204
PTFE (polytetrafluoroethylene) e.g. Teflon [1]			386	998	2.1	2.2	—	0.21	-200	260	320	340
PVF (polyvinyl fluoride) e.g. Tedlar [4]	58		3	7.5	1.38	1.57	22.56	42.05	-70	100	185	190
PVDF (polyvinylidene fluoride) e.g. Kynar [2,4]			3.4	14	1.76	1.78	—	29.09	-30	135	168	172
Nylon 6 (polyamide) extruded	90	110	2.6		1.13		17.76	59.25			210	220
Nylon 6 (polyamide) biaxially oriented			1.2	2.3	1.16		—	125.06				
Nylon 6/6 (polyamide) molding compound					1.13	1.15	—	—			255	265
Nylon 11	323		34	92	1.02		5.48	5.02			191	194
Nylon 12	323		34	92	1.04		5.38	4.92			160	209
Polycarbonate	1600		300		1.2		0.94	0.48				
PET (polyester or polyethylene terephthalate)	100		3	6	1.38	1.41	13.08	42.05			245	265
Polyetherimide			37		1.27		—	3.70				
LDPE (low density polyethylene)	1950		500		0.91	0.925	1.02	0.38			98	124
MDPE (medium density polyethylene)	1950		250	535	0.926	0.94	1.00	0.75			98	124
HDPE (high density polyethylene)			185		0.941	0.965	—	1.00			130	137
UHMWPE (ultrahigh molecular weight polyethylene)					0.94		—	—			125	135
EVA (ethylene vinyl acetate copolymer)			840		0.924	0.94	—	0.22			103	108
Polyimide (e.g. Kapton)	250		25		1.42		5.09	4.90				
PMMA (polymethyl methacrylate)					1.14	1.26	—	—				
Polypropylene extrusion cast	1700		150	240	0.885	0.905	1.20	1.31			160	175
Polypropylene biaxially oriented			100	160	0.902	0.907	—	1.93				
Polystyrene			250	350	1.05	1.06	—	0.66			74	110
Polyurethane elastomer			75	327	1.11	1.24	—	2.09			75	137
PVC (polyvinyl chloride) nonplasticized	3	10	4	30	1.2	1.5	501.58	36.27				
PVC (polyvinyl chloride) plasticized			100	1400	1.2	1.4	—	1.45				
Polysulfone			90	230	1.24	1.37	—	1.56				
TFE/HFP/VDF terpolymer e.g. THV [4,5]			225		1.97	2.03	—	0.39			115	180
Vinyl chloride acetate copolymer nonplasticized			15	20	1.3	1.4	—	8.93				
Vinyl chloride acetate copolymer plasticized			20	150	1.2	1.35	—	7.25				
Vinylidene chloride vinyl chloride copolymer	4	55	0.08	1.1	1.59	1.71	283.92	1368.59				
Vinyl nitrile rubber alloy			50	65	1.1	1.3	—	3.17				

[1] Permeability & temp data from H. Fitz, "Fluorocarbon films-present situation & future outlook", Kunststoffe with German Plastics, vol 70(1) English trans. pp.11-16, Jan(1980).
 [2] Lo O2 permeability data from [1].
 [3] Use permeability data for air (O2 data not available) from [1].
 [4] Use additional permeability data from Allied Signa Data Sheet "Fluoropolymer Barrier Properties", 12/4/95
 [5] 3M Product Specifications including Form 33754-A-PWO December 13, 1993

The LLNL effort to develop the next generation of advanced hydrogen tankage was able to take advantage of existing SRI capability and further a broader understanding of hydrogen permeability. Thiokol has internally funded a new facility at SRI capable of permeation testing at

high pressure (up to 5000 psi or 34.5 MPa) with hydrogen, as well as an unprecedented fixturing arrangement that enabled the first-ever collection of permeation test data under controllable biaxial strain (at the levels anticipated in tank liner service). This test capability has been made available with Thiokol's permission for LLNL research, and is illustrated in Figure 8. LLNL took advantage of an earlier hiatus in Thiokol's testing to procure ASTM-traceable calibration for all subsequent measurements, confirming previous measurements made at LLNL on LLNL-developed liners. LLNL, the USAF, and Thiokol IR&D funded significant additional hydrogen permeation testing on a variety of candidate liner materials, as a function of pressure. The graph in Figure 9 not only confirms the hydrogen permeability of several previously employed liner materials, it extends the sparse earlier results to a much wider range of pressures, temperatures, and materials. This new database, and the literature survey summarized in Table 3 have been used to assess many relevant materials' acceptability as thin liners for high pressure tankage. Downselection is currently proceeding in parallel with rotational molding process development to produce LLNL's next generation of advanced liners (sufficient to enable DOE 2000 Goals).

Tests with gas P up to 5 ksi (34.5 MPa) as a function of temperature & biaxial strain

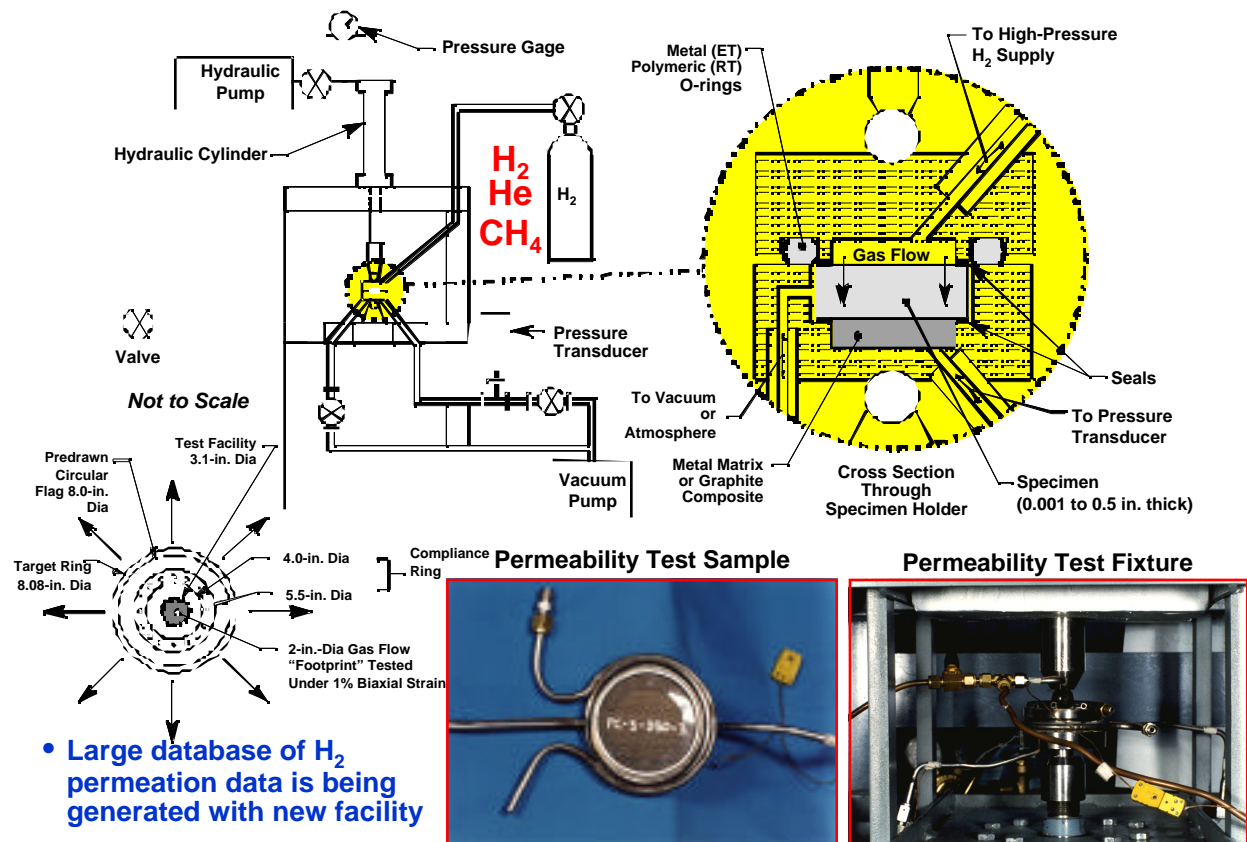


Figure 8 - Schematic and Photos of Permeability Test Fixture at SRI

SRI Measured the Permeability of Several Candidate H₂ Containment Materials as a Function of H₂ Delta Pressure

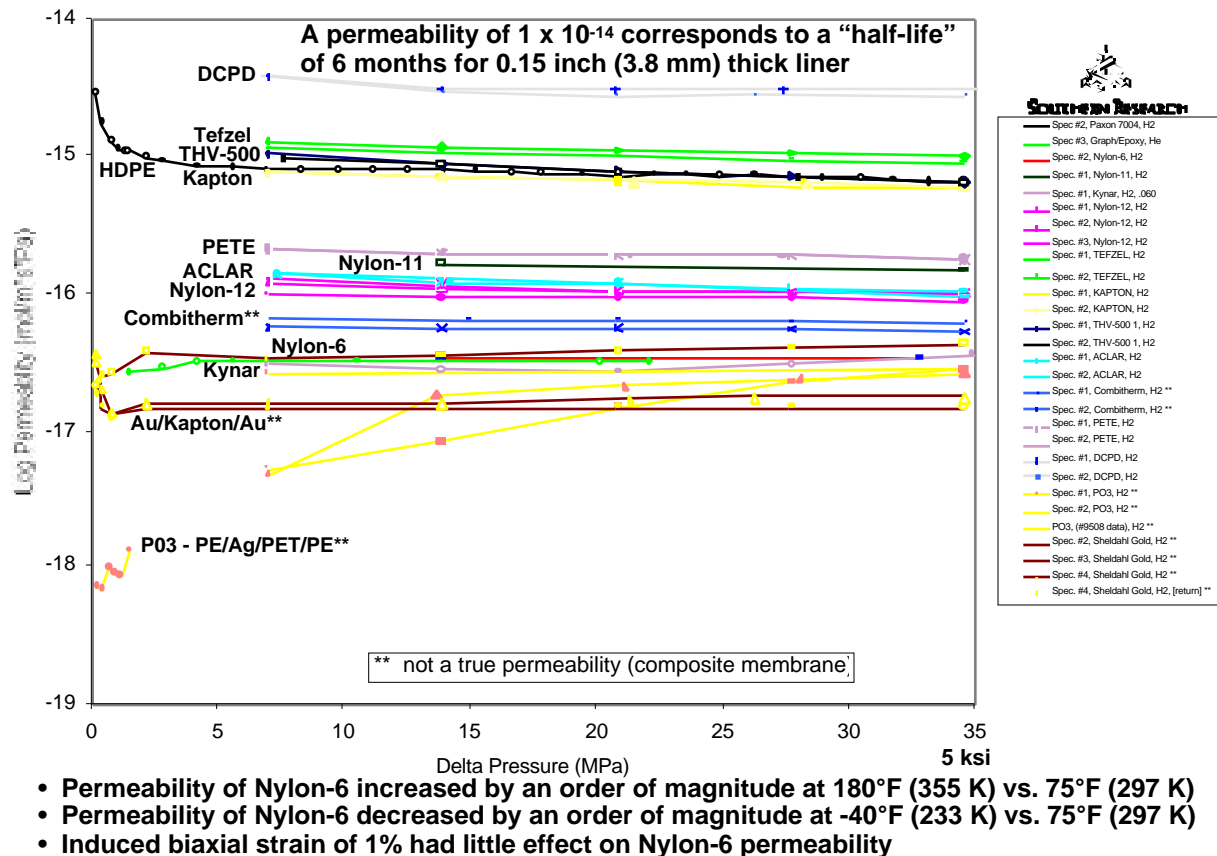


Figure 9 - Measured Hydrogen Permeability of Several Candidate Liner Materials

Past Results

LLNL first attempted to fabricate thin tank liners (bladders) with a proprietary low permeability laminate joined to thick end domes in 1995. These early thick liner domes were fabricated by rotational molding, and their manufacture incorporated bosses molded in place. These early, very thin sidewall bladders were used as inflatable integral mandrels for fabricating lightweight pressure vessels. LLNL's initial prototype vessels had estimated performance factors ($P_b * V / W$) of 40 km (1.6 million inches).

Besides assisting the DOE in formulating a sensible competition for tankage development activities, LLNL has been engaged in close technical collaboration and monitoring of the ensuing Thiokol contract (Golde 1999). This collaboration has enabled the development of a new generation of liners that are readily adopted by Thiokol due to the development work at Aero Tec Laboratories (ATL), under LLNL direct sponsorship, assisted by permeation measurements at Southern Research Institute (SRI).

These past years' tankage developments have enabled energy storage systems designed for high altitude solar rechargeable aircraft and spacecraft to be advantageously adapted to automotive use. A primary fuel cell (FC) test rig was refurbished and converted into a uniquely capable URFC test rig at LLNL, in order to integrate entire energy storage subsystems based on the potential breakthrough specific energy performance such tankage can provide. This rig accomplished LLNL's groundbreaking result of FY1997: a URFC cell was tested using H₂/O₂ for >2000 cycles and showed < a few percent degradation as both fuel cell & electrolyzer!

Subsequent years' work has demonstrated high performance operation (>1000 ASF) of URFCs, for operation in fuel cell and cathode feed electrolyzer modes (Mitlitsky 1998-b, Mitlitsky 1998-e, Mitlitsky 1998-g). High performance operation (>2000 ASF) has also been reported for an operating URFC in FC mode with 1 mg/cm² catalyst loading. This argues strongly that URFCs should not be dismissed on economic grounds, compared to primary fuel cells which have been pursuing reduced catalyst loading possibilities for several years. LLNL URFC performance demonstrations were extended to include a URFC operated on H₂/O₂ in fuel cell mode at 0.6 V to >1000 ASF (>1.1 A/cm²) with 1 mg/cm² catalyst loading in FY1998 (Mitlitsky 1998-b, Mitlitsky 1998-e).

In FY1998 LLNL's URFC test rig was upgraded to accommodate anode feed electrolysis, as well as its previous capability to measure cathode feed electrolysis and FC modes of operation. At roughly the same time, the LLNL team demonstrated rapid cycling (<60 s electrolysis/FC cycle) of a URFC, allowing the accumulation of copious cycle life data that turned out to be enabling for many energy storage applications. The LLNL team also negotiated and put in place agreements to test Hamilton Standard and Proton Energy Systems proprietary hardware on loan to LLNL

Two patents were awarded to LLNL energy storage team members in 1998: F. Mitlitsky, B. Myers, and F. Magnotta, "Lightweight bladder lined pressure vessels," *Disclosure and Record of Invention*, DOE Case No. IL-9722, U.S. Patent No. 5,798,156, August 25 (1998) and F. Mitlitsky, J.B. Truher, J.L. Kaschmitter, and N.J. Colella, "Fabrication of polycrystalline thin films by pulsed laser processing," *Disclosure and Record of Invention*, DOE Case No. IL-9123, U.S. Patent No. 5,714,404, February 3 (1998).

Current Year Results

During the current fiscal year, and in the preceding months since the FY1999 Program Review, this same LLNL team designed a tank which holds ~8.5 lb (3.9 kg) H₂. This design is the first feasible instance of compressed hydrogen storage that attains the DOE 2000 goals of Pb*V/W ~1.85 million inches (~12% H₂ by weight). A new generation of liners and tanks are being developed under contract with ATL, SRI, and Thiokol to reach this important level of performance. Figure 10 shows several pictures of bladders and tanks that have been developed by LLNL and its industrial partners. The top/left set of four pictures shows the early prototype fabricated in FY1996 (with estimated performance factor of 1.6 million inches) which used a bladder with thin laminate that was joined to thick end domes and acted as an inflatable integral mandrel for composite overwrap. The center left is a collection of fabricated bladders and minimum gauge composite tubes that were funded by the USAF in FY1998. The bottom left photo shows minimum gauge rotomolded liners and a composite pressure vessel fabricated using

one such bladder as an inflatable integral mandrel. The photograph on the right in a lightweight (2.3 kg) molded liner that is thin in the cylindrical section and grades to thickened ends as required for the boss detail. Tanks fabricated using molded parts similar to the one shown should be capable of achieving the DOE 2000 goals.



Figure 10 - Photos of LLNL/Thiokol/ATL Bladders and Tanks

Close collaboration and technical management of Thiokol has captured a significant understanding of the issues that will govern successful liner development. The DOE tankage research and development is beginning to furnish the first results of this collaboration in the form of Thiokol's first generation of conformable hydrogen tankage. The fruits of this research and development activity should be available for incorporation into DOE's various hydrogen application demonstrations this year, with certification to the NGV2 safety standard. These conformable units will be ideal for retrofitting existing vehicle designs to storing compressed hydrogen, while LLNL's directly developed DOE 2000 tanks should give "ground up" vehicle designs sufficient range to compete with existing and alternative vehicles.

In the course of this collaboration, LLNL has deliberately acquired a large database of permeability properties for polymers and laminates at pressure differentials up to 5 ksi (34.5 MPa). This database is augmented with permeability properties at varying temperatures and induced biaxial strains (with varying pressure as a parameter). This database is currently being downselected in parallel with actual liner fabrication. A mold has been designed and manufactured

for rapid prototype of lightweight liners with graded thickness, and a new generation of liners commenced fabrication using the new mold in time for DOE's 1999 Annual Program Review.

LLNL has also built and successfully fielded a demonstration of solar powered PEM electrolytic generation of pressurized H_2/O_2 combusted in a rocket engine. This is relevant to DOE interest in source-leveling alternative energy supplies, as one of the first solar to PEM to hydrogen to controlled energy release end-to-end demonstrations performed outside laboratory conditions. Other recent activities that matter to DOE and may be relevant to the readers of this paper include the recent commencement of technical management for the DOE's Renewable H_2 Utility System programs involving SRT's H_2 /halogen project and Proton's PEM electrolyzer project. Proton has recently coupled a PEM hydrogen generator to a SunDishTM system that generates electricity during daytime operation by concentrating solar energy into a Stirling external heat engine (Friedland 1999).

Plans for Future Work

The remainder of FY1999 is likely to encompass the design, fabrication, and testing of tank liners that have improved temperature range and stress/strain characteristics. LLNL will support DOE programs for development of conformable tanks, renewable H_2 utility systems, and regenerative fuel cell systems with assertive technical management of its industrial partners. If funds permit the recertification of LLNL's URFC Test Rig (currently shut down awaiting electrical rebuilding to satisfy new safety concerns, the LLNL team hopes to test alternative PEM membranes and catalyst mixtures for performance improvement of URFCs using H_2/O_2 , H_2 /air, and/or H_2 /halogen chemistries.

In the coming fiscal year (FY2000), the LLNL team expects to work closely with industry to improve the cycle life and certify pressure vessel standards compliance of its directly developed lightweight pressure vessels with advanced liner materials. This process should confirm the achievement of the DOE 200 Goals, along with their implications for the range of future compressed hydrogen fueled vehicles. This team hopes to work with a car company that is willing to adopt DOE tankage technologies to further persuade the alternative fueled vehicles community of the superiority of its hydrogen storage. The team also hopes to once again take advantage of the electrolyzer and URFC Test Facility at LLNL to evaluate industrial PEM hardware and provide systems integration research and development.

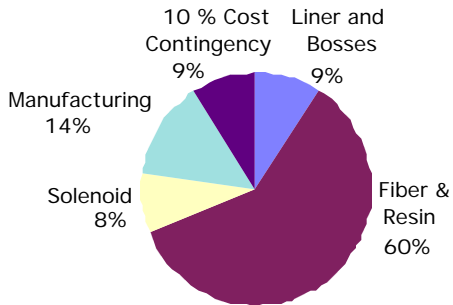
Status of Economic Evaluation/System Analysis

As the DOE has requested, the LLNL team has produced economic evaluations of the vehicular hydrogen storage technology it is advocating in conjunction with Thiokol Corporation, Directed Technologies Inc. (DTI), Toray Composites, and colleagues at LLNL. The major cost driver with the most uncertainty is the cost of various grades of carbon fiber that may be achievable with high volume production. Aggressive estimations for cost of high volume production of T1000G (currently the highest strength-to-weight carbon fiber commercially available) suggest that its cost can be reduced several-fold from its current cost of ~\$70/lb (\$154/kg). Figure 11 summarizes the results of this spreadsheet based economic analysis, and shows some of the graphics that have been used to present economic performance projections.



Economic Evaluation/System Analysis for High Volume Production of Carbon Fiber Tanks

Pressure Vessel Total Cost



Direct H2 Fuel Tank Subsystem

3.58 kg (7.9 lb) of H2 at 34.5 MPa (5,000 psi) stored in carbon fiber tank, integral solenoid and pressure relief device (PRD)

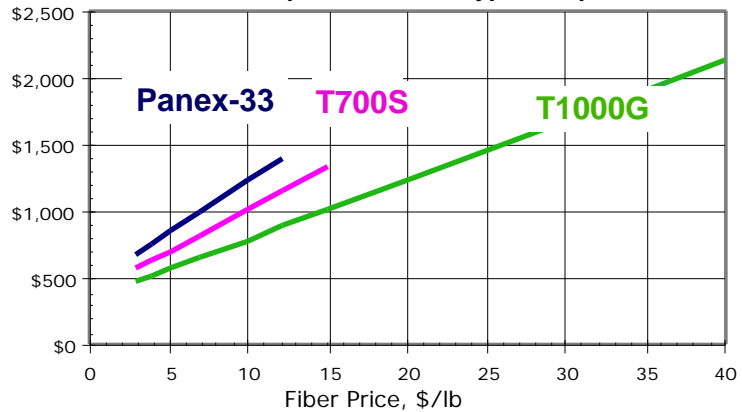
Liner and Bosses	\$78
Fiber & Resin	\$500
Solenoid	\$69
Manufacturing	\$117
10% Cost Contingency	\$76

Tank System Cost \$841

Major Assumptions

- Use Zoltec Panex-33 carbon fiber with manufacturer projected year 2000 price of \$5/lb
- Pressure vessels uses HDPE liner
- High production volume manufacturing (500,000 units/year)

Tank System Cost (pressure vessel & solenoid) as a function of fiber price with fiber type as a parameter



Fiber Type	Current Price \$/lb	Projected Price \$/lb
Panex-33	8	5-6
T700S	15-21	12-15
T1000G	70	15-25

- Tank System Cost of \$841 corresponds to \$6.93/kWh vs DOE 2000 goal of \$20/kWh
- Tank System Cost of \$1015 corresponds to \$8.24/kWh using similar assumptions for T1000G with a future price of \$15/lb

Figure 11 - Economic Evaluation/System Analysis for High Volume Tank Production

Although the use of T1000G will result in the lightest weight tanks, its cost premium must be compared to lower strength/less expensive carbon fibers, such as M30SC with current cost of ~\$30/lb (\$66/kg), or to even lower strength/less expensive fibers, such as T700S with current cost of \$15-21/lb (\$33-46/kg) or to Panex-33 with current cost of ~\$8/lb (\$18/kg). High volume cost projections for these fibers (500,000 units/yr) have been estimated by DTI (with input from manufacturers) to be \$15-25/lb (\$33-55/kg) for T1000G, \$12-15/lb (\$26-33/kg) for T700S, and \$5-6/lb (\$11-13/kg) for Panex-33. M30SC would have an estimated high volume cost of \$15-20/lb (\$33-44/kg).

Assuming materials choices of Panex-33 at \$5/lb (\$11/kg) with a high density polyethylene (HDPE) liner, the cost of a tank that is capable of storing 8.0 lb (3.6 kg) of hydrogen has been estimated by DTI to be \$841 (\$78 for liner and bosses, \$500 for fiber & resin, \$69 for solenoid, \$117 for manufacturing, and a 10% cost contingency). These assumptions for Panex-33 result in cost estimates of \$105/lb (\$231/kg) of hydrogen stored or \$6.93/kWh. Note that by repeating this analysis for a tank with similar capacity using T1000G, assuming a fiber cost of \$15/lb (\$33/kg), the estimated tank cost in high volume production would be ~\$1000/unit (~\$125/lb or ~\$275/kg of hydrogen stored or ~\$8.24/kWh). This estimate beats the DOE 2000 goal of \$20/kWh, for a tank that can meet the technical performance rigors of DOE 2000 Goals. The 3-fold increase in

the price of T1000G fiber compared to Panex-33 fiber is largely offset with significantly less fiber and resin, in addition to marginal decrease in manufacturing cost. (Note that the tank made with Panex-33 would be ~75% heavier than the tank made with T1000G.) The above calculations are based on aggressive assumptions for high volume fiber costs and for projected material properties of Panex-33.

Goals and Basis for Goals

The main thrust of the LLNL team's work in FY1999 has focused on close collaboration with industrial partners to design, fabricate, and test moldable polymer liners which have improved properties compared to high density polyethylene (HDPE). Continuing work with industrial partners should reduce the liner mass of moldable polymer liners even further. Prototype tanks should be tested in integrated leak tests to verify their adequately low permeation at ~1% strain. This activity should complete the demonstration of a prototype tank design that is capable of storing 8-8.5 lb (3.6-3.9 kg) hydrogen at 5000 psi (34.5 MPa) with >10% hydrogen by weight (mass H₂ / mass [H₂ + tank]). If successful, this direct attempt will meet several of the DOE 2000 targets in a series of prototypes capable of 700 Wh/liter, specific energy of 4,000 Wh/kg, and 12% H₂ by weight. At least one of the fabricated demonstration tanks will be burst to begin the long and costly process that would lead this technology to full hydrogen pressure vessel safety certification (when funds are available).

When URFC testing at LLNL is funded again, the team will work to achieve the DOE fuel cell performance goals (>1.1 A/cm² (1000 ASF) @ 0.6 V in fuel cell mode for a single cell URFC) using H₂/air and/or reduced catalyst loading. A significant improvement in test rig construction methods (in order to attain "touch safe" certification) has been required by changes in LLNL inspection procedures, and all new requirements are being speedily addressed (as funds permit). The LLNL URFC test rig is expected to be re-certified and back on line shortly, enabling the independent confirmation of Proton's claims about their URFC technology (with high catalyst loading) and/or testing of URFC MEAs fabricated by Hamilton Standard Space & Sea Systems with catalyst loading from 0.25 - 4 mg/cm² on Nafion 105. The team will certainly exert close technical supervision of Proton and Thiokol, as well as commencing the technical supervision of SRT.

In FY2000 the LLNL energy storage team expects to demonstrate adequate temperature, pressure, and humidity cycle life of this year's prototype tanks. Funding should be sufficient to fabricate a sufficient quantity (likely 5) of prototype tanks to allow a statistically-significant confirmation of their burst pressure rating. Commencing NGV2 and integrated H₂ leak rate testing will begin the costly process of officially confirming the service worthiness of LLNL's advanced tankage technologies. Continuing tank economics information capture is likely as more tanks are wrapped, tooling is developed, and industrial contacts are made.

The LLNL energy storage team hopes to achieve and surpass URFC performance goals from FY1998 (not funded in FY1999) by testing new cell components with H₂/air and/or further reduced catalyst loading (<1 mg/cm²/electrode). These tests could show that URFCs are viable for high volume applications, where catalyst loading is a significant cost component. Close technical supervision of Proton, Thiokol, and SRT will provide DOE with the best technology,

research, documentation, and value for several ambitious contracts. LLNL's provision for a technically assertive interface between various DOE-funded demonstration projects should also be actively pursued in order to improve the chances that subsystems being developed can actually work in the target vehicles and utility systems the DOE hopes to demonstrate.

Major Barriers to Meeting Goals

The technical superiority of the energy storage systems that LLNL is advocating is not in doubt, but eventual adoption depends on high volume price reductions as well as public acceptance. The cost of carbon fiber is the principal driver for tank costs. Using current performance data, DOE cost targets can be met only with some fairly aggressive assumptions regarding future high volume carbon fiber pricing. The extrapolation of carbon fiber prices over several orders of magnitude in production volume is difficult to predict, since little price forecast information on this recently introduced exotic material is available now.

The carbon fiber cost barrier applies to cylindrical as well as conformable tanks. Although cylindrical tanks could provide lighter weight, less expensive storage, the design traditions and existing practices/tooling/workforce puts significant burdens on the kind of "ground up" vehicle design that could benefit from single large cylinders. Figure 12 illustrates the problem of packing 3.6 kg of hydrogen at 5000 psi into a passenger car, if not designed in from the ground up. A single large cylinder (~46 cm OD x 122 cm long) can be placed under a raised rear seat or between split front seats. Alternatively a ~30 cm OD x ~270 cm long tank can run the length of the car. If the OD is kept to ~30 cm or less, packaging can be done into a sandwich floor construction, like that found in the A-Class configuration. Besides finding the room for both tanks and passengers, a complex cluster of safety and regulation issues remain to be resolved before new H₂ mobile and utility technology can be deployed in widespread applications. Technical superiority of the alternatives that the LLNL team is developing may only win out over reduced development costs once a market of significant size has opened up for this class of vehicle.

Vehicles with URFC energy storage are cost competitive with battery augmented primary fuel cell powered vehicles and have the critical bootstrapping feature of carrying a hydrogen infrastructure where the vehicle is driven. However, URFCs embody substantial intellectual property and are currently available from few manufacturers. Experience with utility applications could well change this situation, at least at a larger (> 50 kW) scale, yet operating experience with energy storage systems built around electrochemical cell stacks is almost non-existent today, closely held by a few experts, and generally decades old. Many components suitable for cost-effective systems must be modified or re-engineered entirely from available parts designed for other (primarily aerospace) uses. Less importantly, recent advances in electrochemical membranes and catalysts for primary fuel cells have not yet been applied to URFCs. URFCs have yet to be combined with lightweight ancillaries (including advanced pressure vessels) capable of delivering breakthroughs in vehicular energy storage, so even critical systems' performance demonstrations may be a few years away. Enroute to those demonstrations, control, operations, and experimental procedures must be learned and perfected as system integration progresses. As is manifestly true for their key components, cost reductions will be required to make such systems economically competitive in almost every (including aerospace) application.

Single Cylinder is Lowest Tankage System Cost & Complexity but Requires Car to be Designed from the Ground Up with that Priority

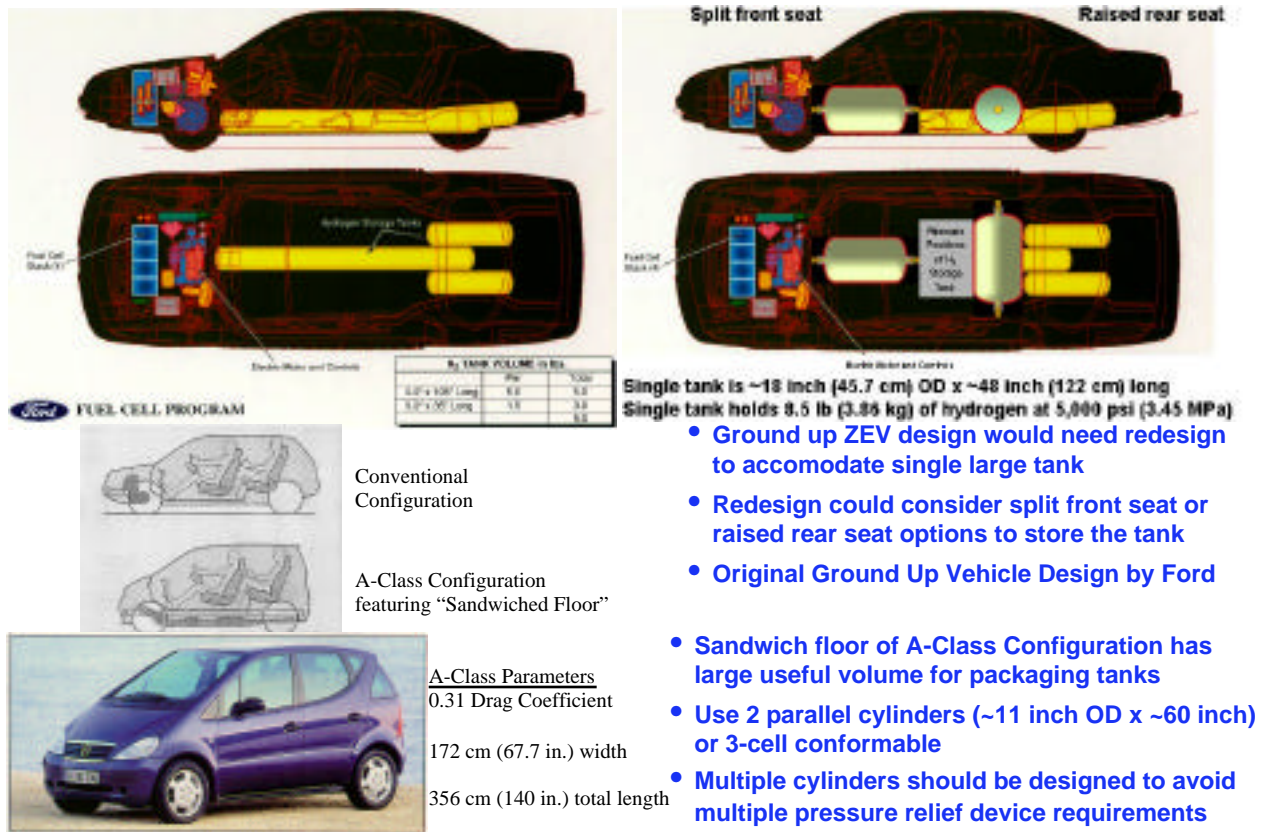


Figure 12 - Tank Size and Location Considerations for Vehicular H₂ Storage

Conclusions

URFC systems with lightweight pressure vessels have been designed for automobiles. These energy storage systems are expected to be cost competitive with primary FC powered vehicles operating on hydrogen/air with capacitors or batteries for power peaking and regenerative braking (Mitlitsky 1999-b, Mitlitsky 1998-b, Mitlitsky 1994). URFC powered vehicles can be safely and rapidly (< 5 minutes) refueled from high pressure hydrogen sources, when available, to achieve driving ranges in excess of 380 miles (610 km). The employment of URFCs would save the consumer the entire capital cost of a home hydrogen generation unit. That consumer would be able to electrically recharge at any available electrical source, instead of being tethered to a single home electrolysis unit. URFC-powered automobiles would still be able to rapidly refuel by direct hydrogen transfer when a hydrogen infrastructure becomes available.

Whether electrically refuelable or not, a vehicle powered by compressed H₂ (at 5,000 psi) is now the system to beat, as DTI has concluded (James 1999). Such a vehicle offers: Low Weight (>10% H₂ fraction) while storing hydrogen in an acceptable volume, at an acceptable cost. Its other advantages include high system simplicity, high safety, the potential for faster refills than

its competitors, as well as expected support by a feasible H₂ infrastructure (in both start-up and mature phases).

Acknowledgments

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References

- Aceves, S.M., J. Martinez-Frias, and O. Garcia-Villazana. 1999. "Analysis and experimental testing of insulated pressure vessels for automotive hydrogen storage," (in these proceedings).
- Appleby, A.J. and E.B. Yeager. 1986. *Energy*, 1986, 11, 137.
- Arthur D. Little, Inc. 1994. "Multi-fuel reformers for fuel cells used in transportation: Assessment of hydrogen storage technologies," Report Number DOE/CE/50343-1, March 1994.
- Beaufriere, A., R.S. Yeo, S. Srinivasan, J. McElroy, and G. Hart. 1977. *12th IECEC*, 1977, 959-963.
- Carter II, P.H., F. Mitlitsky, A.H. Weisberg, J.C. Whitehead, and R.W. Humble. 1998. "Design trade space for a Mars ascent vehicle for a Mars sample return mission," IAA Conference on Low-Cost Planetary Missions, Pasadena, CA, April 27-May 1, 1998; UCRL-JC-130277.
- Chludzinski, P.J., I.F. Danzig, A.P. Fickett, and D.W. Craft. 1973. General Electric Company Technical Report AFAPL-TR-73-34, June 1973.
- Daney, D.E., F.J. Edeskuty, M.A. Daugherty, F.C. Prenger, and D.D. Hill. 1995. *Cryogenic Engineering Conference*, July 17-21, 1995.
- de Groot, W.A., L.A. Arrington, J.F. McElroy, F. Mitlitsky, A.H. Weisberg, P.H. Carter II, B. Myers, and B.D. Reed. 1997. "Electrolysis propulsion for spacecraft applications," AIAA 97-2948, 33rd AIAA/ASME/SAE/ASEE Joint Propulsion Conference, July 7-9, 1997.
- Friedland, R.J., A.J. Speranza. 1999. "Integrated renewable hydrogen utility system," (in these proceedings).
- Golde, R., B. McQuivey, R.K. Kunz. 1999. "Conformable hydrogen storage development," (in these proceedings).
- James, B.D., C.E. Thomas, and F.D. Lomax, Jr. 1999. Directed Technologies, Inc., "Onboard Compressed Hydrogen Storage," presented at the 1999 Canadian Hydrogen Association Meeting, February 1999.
- Jasionowski, W., K.J. Kountz, and C.F. Blazek. 1992. Gas Research Institute Report #GRI-92/0350, July 1992.

Kare, J.T., F. Mitlitsky, and A.H. Weisberg. 1999. "Preliminary demonstration of power beaming with non-coherent laser diode arrays," Space Technology & Applications International Forum (STAIF-99), Albuquerque, NM, January 31-February 4, 1999; UCRL-JC-133428.

Kountz, K.J. 1994. *Proc. 207th ACS-Division of Fuel Chemistry*, March 13-17, 1994.

McElroy, J.F. (Hamilton Standard); R.W. Humble (US Air Force Academy); W.A. de Groot and S.R. Oleson (FDC, Inc., NASA LeRC); and F. Mitlitsky, A.H. Weisberg, B. Myers, and P.H. Carter II (LLNL). 1998. "Unitized Electrolysis Propulsion and Fuel Cell Power for Selected Satellite Missions," *33rd Intersociety Energy Conversion Engineering Conference (IECEC)*, Colorado Springs, CO, August 2-6, 1998 (IECEC-98-339).

McElroy, J. and G.G. Patwa. 1979. *Proc. 72nd AIChE*, Nov. 25-29, 1979.

McElroy, J. 1977. *Proc. DOE Chemical Energy Storage and Hydrogen Energy Systems Contract Review*, JPL Publication 78-1, November 16-17, 1977, pp. 27-33.

Mitlitsky, F., B. Myers, A.H. Weisberg, and A. Leonida. 1999-a. "Applications and Development of High Pressure PEM Systems," Portable Fuel Cells, Lucerne, Switzerland, June 21-24, 1999.

Mitlitsky, F., B. Myers, A.H. Weisberg, T.M. Molter, and W.F. Smith. 1999-b. "Reversible (Unitized) PEM Fuel Cell Devices," Portable Fuel Cells, Lucerne, Switzerland, June 21-24, 1999.

Mitlitsky, F. 1998-a. "Water Rocket Technologies," invited presentation, US Embassy, Paris, France, December 10-11, 1998; UCRL-MI-132628 Rev 1.

Mitlitsky, F., B. Myers, A.H. Weisberg, and T.M. Molter. 1998-b. "Unitized regenerative fuel cell systems," *1998 Fuel Cell Seminar*, Palm Springs, CA, November 16-19, 1998; UCRL-JC-130198 (paper) and UCRL-MI-130198 (viewgraphs).

Mitlitsky, F., P.H. Carter, M.D. Dittman, R.W. Humble, J.T. Kare, B. Myers, and A.H. Weisberg. 1998-c "Water Rocket Design Trade Analysis," whitepaper to Air Force Research Lab, October 29, 1998.

Mitlitsky, F. 1998-d. "Regenerative Fuel Cell Systems," Invited Presentation, Joint DOE/EPRI/GRI Workshop on Fuel Cell Technology, San Francisco, CA, May 18-20, 1998.

Mitlitsky, F., B. Myers, and A.H. Weisberg. 1998-e. "Regenerative Fuel Cell Systems R&D," DOE Annual H₂ Program Peer Review, Alexandria, VA, April 28-30 (1998); UCRL-JC-131087.

Mitlitsky, F., and J.B. O'Sullivan (contributors), T.M. Molter, R.H. Byron, M.E. Dristy, and J.K. Shiepe. 1998-f. "PEM Unitized regenerative fuel cells for integrated renewable hydrogen energy storage," 9th Annual U.S. Hydrogen Meeting, National Hydrogen Association, Vienna, VA, March 4, 1998.

Mitlitsky, F., A.H. Weisberg, and B. Myers. 1998-g. "Regenerative fuel cell systems," *Energy & Fuels*, 12 (1), 56-71, January 12, 1998; UCRL-JC-128267 Rev 1;
<http://pubs.acs.org/subscribe/journals/enfuem/jtext.cgi?enfuem/12/i01/html/ef970151w.html>

Mitlitsky, F., B. Myers, and A.H. Weisberg. 1996-a. "Lightweight pressure vessels and unitized regenerative fuel cells," *1996 Fuel Cell Seminar*, Orlando, FL, November 17-20, 1996; UCRL-JC-125220 (paper) and UCRL-MI-125220 (viewgraphs).

Mitlitsky, F., W.A. de Groot, L. Butler, and J.F. McElroy. 1996-b. "Integrated modular propulsion and regenerative electro-energy storage system (IMPRESS) for small satellites," *AIAA Small Satellite Conference*, September 16-20, 1996; UCRL-JC-125242.

Mitlitsky, F. and B. Myers. 1996-c. "Development of an advanced, composite, lightweight, high pressure storage tank for on-board storage of compressed hydrogen," *Fuel Cells for*

Transportation TOPTEC: Addressing the Fuel Infrastructure Issue, Alexandria, VA, April 1-2, 1996; UCRL-MI-123802.

Mitlitsky, F. 1996-d. "Rechargeable solar powered aircraft using regenerative fuel cells," *High Altitude Long Endurance (HALE) Roundtable Meeting, European Space Research and Technology Centre (ESTEC)*, invited presentation, Noordwijk, The Netherlands, January 17, 1996; UCRL-MI-122808.

Mitlitsky, F., N.J. Colella, and B. Myers. 1994. "Unitized Regenerative Fuel Cells for Solar Rechargeable Aircraft and Zero Emission Vehicles," *1994 Fuel Cell Seminar*, November 28-December 1, 1994; UCRL-JC-117130 (paper) and UCRL-MI-117130 (viewgraphs).

Mitlitsky, F., N.J. Colella, B. Myers, and C.J. Anderson. 1993. "Regenerative fuel cells for solar powered aircraft," *28th Intersociety Energy Conversion Engineering Conference (IECEC)*, August 9-13, 1993; UCRL-JC-113485.

NASA Dryden Press Release 98-64. 1998. August 7, 1998;
<http://www.dfrc.nasa.gov/PAO/PressReleases/1998/98-64.html>

Nutall, L. J. 1982. "Development progress on solid polymer electrolyte fuel cells with various reactant sources," General Electric, Electrochemical Energy Conversion Programs, Report.

Souers, P.C. 1986. *Hydrogen Properties for Fusion Energy*, University of California Press: Berkeley, CA, pp 370-373.

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