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Technical and Systems Evaluations

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Abstract

During FY 1998 Energetics performed a variety of technology-based evaluations for the Hydrogen Program. Three evaluations are summarized below: hydrogen bromine-based electricity storage, carbon-based hydrogen storage, and hydrogen-fueled buses.

Hydrogen Bromine-based Electricity Storage

The hydrogen bromine electrochemical cycle, shown in reaction {1} below, has been previously recommended as a means of storing electricity because, unlike the water cycle, it is highly reversible with demonstrated electric-to-electric efficiencies of 80% or higher (Fritts and Savinell 1983, Balko and McElroy 1980). The HBr-based electricity storage system is best suited for 6-10 hour peak shaving applications and is envisioned as a part of a distributed generation system.

$$H_2 + Br_2 \longleftrightarrow 2\,HBr \qquad \{1\}$$

Figure 1 is a schematic of the proposed HBr-based electricity storage system. The heart of the system is the reversible PEM cell, which can be operated in either electrolysis or galvanic mode. Because the HBr reaction is highly reversible, the electrode catalysts for electrolysis and galvanic operation are not highly specialized, making it less challenging to fabricate a reversible cell that is efficient in both directions.

The process operation is as follows. HBr is electrolyzed during periods of low electricity demand, forming bromine liquid and hydrogen gas which are stored. Hydrogen gas and bromine are recombined in the cell during peak demand hours to produce electricity. A power electronics component rectifies/inverts electricity in and out of the electrochemical cell and manages the voltage difference between the stack and the load.

This HBr technology is being developed by SRT Group and National Power, who have developed solar-based HBr energy systems and sodium bromide electrochemical energy storage systems, respectively. The hydrogen bromine electricity storage is in the pre-commercial stage and faces the following technical challenges:

> Previous laboratory scale demonstrations of HBr cell efficiencies were conducted at low current density (i.e., less than 200 Amps/ft^2). High efficiency must be demonstrated at a current density above 500 Amps/ft^2.
>
> Because hydrobromic acid is electrically conductive, shunt currents limit the number of cells that can be stacked in a bipolar configuration. Modular stack output potential is limited to 10-20 volts. Such low voltage will create high resistive losses and increase the cost of power electronics equipment, which is generally current-limited. Several stacks could be externally wired in series, but this increases the cost of the system as well. An optimal system that considers stack voltage limitation must be developed.
>
> The field life of the membrane electrode assemblies (MEAs) must be demonstrated. Because of the toxicity of the reactants, changing out the MEAs could be expensive compared to a water system. We project that an MEA life of 5-7 years is required for an economic system.
>
> The cost of the hydrogen compressor is based on the assumption that the gas outlet pressure from the PEM stack is 1,000 psi. In theory, a high hydrogen gas outlet pressure is achievable at low cost by pressuring the water inlet. If only a 200 psi outlet pressure is achievable, the cost of the hydrogen compressor would double.

Table 1 shows a capital cost estimate for a 300 kW 8 hour HBr-based electricity storage system. The estimate is based on a 72% AC/AC round-trip efficiency, a reversible PEM stack cost of 350 $/kW, and a power electronics cost of roughly 175 $/kW. All these estimates were provided by SRT Group and National Power and inherently rely on the resolution of the above technical issues.

A financial analysis of the system was conducted based on a hypothetical commercial or industrial facility that faces a time-of-use electric rate structure and buys an HBr system to reduce its electric bill. The HBr system would be operated 8 hours per day, 250 days per year. A pro forma cash flow analysis model was used to determine the energy rate for peak electricity required to yield a 15% after-tax internal rate of return based on an off-peak electric rate of 2 cent/kWh and a demand charge of 8 $/kW/month (financial analysis assumptions include 20 year system life, 50% debt/equity, 7.5% interest on debt, 10 year ACRS depreciation schedule and 28% marginal tax rate). The base case peak electricity cost is 7.9 cents/kWh. Figure 2 shows that the required cost of peak electricity varies between 6 and 12 cents/kWh based on changes in several key assumptions.

Table 1. Capital Cost Estimate for a 300 kW, 8 hour Hydrogen Bromine-based Electricity Storage System

Component	Cost
PEM stack (300 kW, 350 $/kW)	105,000
Power electronics (inverter 125 $/kW, rectifier 50 $/kW)	55,000
Hydrogen Compressor (10 kW, 12.6 kg H_2 per hr)	26,300
Hydrogen gas storage (84 kg, 400 $/kg)	33,800
Acid storage tank (Kynar, 10.7 m^3, 6,000 $/$m^3$ installed)	64,000
Piping and controls	30,000
Assembly and installation	30,000
Total capital cost	**$ 344,100**
Size of equipment based on a 72% round trip efficiency	

As a comparison, a diesel generator (12 kWh/gallon efficiency, 1 $/gallon fuel cost, 1 cent/kWh O&M) operated 8 hrs/day 250 days per year requires an electricity rate of 5.4 cents/kWh to yield a 15% IRR. On the other hand, the 1996 average rate for electricity for industrial customers in New England was 7.92 cents/kWh (both peak and off-peak, *Electric Power Annual* 1996 Volume II, 1997). Our assessment is that the HBr-based electricity storage system has the potential to be commercially viable if the developers achieve their cost and efficiency goals.

A proposed area of future work is to evaluate the HBr system as an addition to wind/diesel hybrid systems in remote applications. The HBr system would manage the interface among the wind turbine generators, the diesel generators, and the load. It would enable the diesel generators to run at a more level rate increasing the fuel efficiency and extending their useful life. Lead acid batteries have been proposed to serve the same function. The HBr system offers the benefit that longer duration storage is less costly. A deeper storage system can capture a significant amount of excess turbine energy and allow a higher capacity wind turbine system for a given size load.

Carbon-based Hydrogen Storage

Carbon-based storage systems use carbon-based structures as a matrix for reversibly binding hydrogen either chemically or physically, providing a hydrogen energy storage medium. Carbon is an attractive material for such a storage system in that it is relatively light and relatively inexpensive. These are two qualities of great importance toward a goal of being able to provide a cost-effective, efficient, on-board storage system for hydrogen. Figure 3 compares the gravimetric and volumetric loading potential of hydrogen in carbon structures with other standard and state-of-the-art hydrogen storage technologies. While some of these other technologies are close to or at the DOE goal for gravimetric loading, most are well short of the volumetric goal. In addition, some storage systems (liquid hydrogen, for instance) involve an energy-intensive production process. This section compares three somewhat different technologies that rely on carbon to store hydrogen. Each is represented by a project that is currently being supported by the U.S. Department of Energy.

1. Carbon Nanotube Materials for Hydrogen Storage - National Renewable Energy Laboratory
2. Hydrogen Storage in Carbon Nanofibers - Northeastern University
3. Thermal Management Technology for Hydrogen Storage - Oak Ridge National Laboratory

Nanotubes are rolled planar structures in tubular form having nanometer-sized diameters, and lengths that are orders of magnitude (micron range) greater than the diameters. The end caps are fullerene-like structures that must be chemically or mechanically removed prior to hydrogen being able to be sorbed into the capillary. Nanofibers have similar dimensions to nanotubes, but are not hollow. Instead, they are stacked plates of graphitic carbon. The hydrogen selectively fits into the spaces between the plates and is adsorbed onto the planar surfaces. The Oak Ridge project uses C_{60} fullerene cage structures as a means to store hydrogen physically, chemically or both. Table 2 provides some comparative production and utilization parameters for the three carbon structures.

Table 2. Comparison of Technologies Using Carbon Structures for Hydrogen Storage

Project	Production Method	Projected weight % Hydrogen	Postulated Sorption Mechanism	Hydrogenation/ Dehydrogenation Temp	Hydrogenation/ Dehydrogenation Pressure
Nanotubes/ NREL	Arc-Discharge or Laser Vaporization	5-10	Adsorption on inner walls of nanotubes	Room Temp.	Currently 15 psi; (projected to be up to a few hundred psi to fill long tubes)
Nanofibers/ Northeastern	Vaporization and Catalytic Condensation	10-75	Several layers of hydrogen selectively condensed between graphite platelets	Near Room Temp. (possibility of needing additional heat to speed sorption process)	About 1500 psi; pressure to release hydrogen
Fullerenes/ ORNL and MER	Arc-furnace	6-8	Physical sorption and/or chemical reaction	200-400°C (lower w/ catalyst)	500 -4000 psi (lower w/ catalyst)

Carbon Nanotubes, NREL

The NREL project has several years of DOE research behind it, as opposed to the other two relatively newer projects. The project has focused on developing methods to produce nanotubes in quantity and on being able to fill the tubes with hydrogen. As a result, NREL pursued various modifications of arc-furnace and laser vaporization nanotube production methods. To date, the arc-furnace method has produced very low yields of nanotubes (less than 0.1 percent). The nanotubes that were present, however, were able to adsorb up to 10 weight percent hydrogen. The laser vaporization method has produced high yields of nanotubes (60-100 percent). These tubes, however, are capped with difficult to remove fullerene-structured ends. (The arc-furnace nanotube ends are much less stable). In addition the laser nanotubes have a very high aspect ratio. These two properties are barriers to hydrogen adsorption. The NREL research team is currently focusing on the nanotubes made by laser vaporization, investigating methods that will cut (and in the same process, decap) the tubes.

Nanotubes hold promise for uniformity and reproducibility. Choosing one of the modes of nanotube production appears to produce bundles of nanotubes at a uniform diameter, and these nanotubes will be aligned parallel to one another due to Vander Waal's forces. This leads to an opportunity for easy packing of nanotube bundles into tanks.

Of the three projects discussed here, the NREL project appears to hold out the promise for the operation which is closest to ambient pressure and temperature conditions.

Fullerene Option, ORNL & MER

The fullerene project at ORNL and MER relies on preparation of so-called "bucky balls," caged structures having a basic formulation of C_{60} or higher, and physically or chemically incorporating hydrogen within the structure. The researchers have demonstrated hydrogenated fullerenes as a bulk powder, pelletized, and deposited in a thin film on aluminum foil. Stable three and four percent by weight hydrogen content has been shown.

If we assume that chemically each carbon, being bonded to three other carbons, can bond with one hydrogen forming $C_{60}H_{60}$, such a system would have a hydrogen weight percent of 7.7, effectively putting an upper limit on the maximum potential hydrogen loading.

An interesting argument can be developed if we look at some of the data coming out of this research project. The ORNL and MER researchers (Wang et al 1996) state that if one looks at the removal rate of hydrogen from the hydrogenated fullerene, where no catalyst is being used, it can be found to obey the expression:

$$\text{Rate} = A\,(C/C_{max})\,e^{-E/\kappa T} \qquad \{2\}$$

where C is the hydrogen concentration in the fullerene and C_{max} is its maximum concentration, A is the Arrhenius pre-exponential factor, E is the activation energy, κ is the molecular gas constant, and T is temperature. Since the rate of a chemical reaction, (dr/dt), is a function of the reactant concentrations and a rate constant, the rate of a reaction

$$aA + bB + \ldots nN \;\text{---->}\; \text{Products} \qquad \{3\}$$

can be expressed as:

$$(dr/dt) = \text{Rate} = k\,[C_A]^a[C_B]^b \ldots [C_N]^n \qquad \{4\}$$

where k is the rate constant and C represents the concentration of the species in question.

The Arrhenius expression for the rate constant, k, can be expressed as a function of temperature by:

$$k = A\,e^{-E/\kappa T} \qquad \{5\}$$

Therefore, the expression for the dehydrogenation process in {2} can also be expressed as:

$$\text{Rate} = k\,(C/C_{max}) \qquad \{6\}$$

This is the form of a first-order reaction, dependent only on the concentration of C, in this case, the concentration of hydrogen. This would indicate that since the reaction rate is not dependent on the rate of concentration of the fullerene hydrogenate, that the fullerene hydrogenate is not a chemical species, but that the hydrogen is physically held within the fullerene structure. At least, it indicates that the rate-determining step is the physical removal of hydrogen from the matrix. This means that the theoretical chemical limit to the degree of hydrogenation may not be the actual limit. It also adds some additional interest to this topic in that there is a possibility that both physical and chemical hydrogen take-up is possible, adding to the potential for hydrogen loading.

Indeed, in some of their later work, the researchers have shown that when a catalyst is used in the fullerene system, it appears that a second rate constant comes into effect (Murphy et al 1997). This may indicate that under these conditions a chemical component has been added to the hydrogen sorption mechanism.

Analysis of the fullerene project is not yet completed, and a visit is planned to MER during this fiscal year.

Carbon Nanofibers, Northeastern University

The Northeastern project is the most controversial of the three. The researchers have presented data indicating that they have attained up to 75 percent hydrogen by weight in a carbon nanofiber matrix. Their initial theoretical data indicated that the graphitic plates that make up the nanofiber are planar on a molecular level, and that hydrogen could adsorb on to these planar surfaces in a complete monolayer. Based on the sizes and packing structure of graphite platelets and hydrogen molecules, this would allow 6.9 liters of hydrogen (STP) to adsorb onto each gram of graphite. This is equal to:

(6.9 liters) x (2 g H_2/ 22.4 liters @STP) = 0.62 g H_2, and

0.62 g H_2 on 1 g carbon = 0.62/(0.62 +1) = 38.1 percent by weight hydrogen.

This is a very lofty claim. However, this number still assumes a perfect monolayer of hydrogen on a perfect planar graphite surface, at the molecular level. One might surmise that a perfect monolayer is unlikely and that the actual amount of coverage of the graphite platelet might be much lower. The researchers, however, claimed that in actuality, the hydrogen take-up was much greater than the monolayer amount, and that in fact, hydrogen was condensing in amounts equivalent to several layers of thickness in between the graphite platelets. Numbers as high as 37 STP liters per gram of carbon (nearly 77 percent hydrogen by weight) were reported informally, and numbers as high as 67 percent hydrogen by weight are reported in the peer-reviewed literature (Chambers et al 1998). Data curves provided by the researchers indicated that exposing the fibers to hydrogen in a closed system at high pressure and room temperature resulted in a pressure drop equivalent to the hydrogen take-up levels claimed, and that exposure to non-nanofiber carbon structures or other materials resulted in little or no pressure drop. The multilayer coverage was attributed to delocalized B electrons associated with the graphite matrix.

The claims here are very great. So are the potential rewards. Inexpensive, safe, on-board storage is important to the use of hydrogen in transportation. If the nanofiber data is indeed accurate, it projects a driving range of up to several thousand miles on a tank of hydrogen (at 10 liters of hydrogen per gram of carbon, a tank 10 gallons in volume would contain enough hydrogen for over 2000 miles of driving).

None of the three carbon-based hydrogen storage projects are close to providing an on-board storage system today, but each has potential. The NREL project will, either by cutting the laser-generated nanotubes or increasing the yield of furnace-generated nanotubes, provide the system that will be able to cycle hydrogen most efficiently, being the closest to ambient conditions (See the Table 2) for temperature and pressure. The fullerene project has demonstrated stable hydrogen containing species with several percent hydrogen present. The fullerenes also require high temperature and pressure conditions to both store and desorb hydrogen. In this way, they are similar to hydrides. The major effort is to catalytically reduce the temperature and pressure requirements. The nanofibers require the most pressurization, and may require some heat to speed the process, but the claimed hydrogen take-up far exceeds all others. The nanotube project is badly in need of independent experimental verification.

Hydrogen-fueled Buses

Table 3 shows significant recent industrial activity aimed at developing hybrid electric power systems for vehicles. A hybrid electric vehicle (HEV) offers improved fuel efficiency, reduced pollutant emissions, and reduced wear and tear on the mechanical components over vehicles powered by a conventional internal combustion engine (ICE). For transit bus applications, hybrid electric vehiles offer adequate range where all-electric vehicles fall short. Most commercial efforts are focused on diesel, gasoline, or natural gas-fueled systems. Energetics has analyzed the feasibility of using hydrogen fuel for hybrid electric power systems and the incremental cost compared to diesel, gasoline, or natural gas-fueled systems. The objective of the analysis is to build on the Hydrogen Program's experience with the Savannah River Bus and explore the impact of more advanced hydrogen storage and hydrogen-fueled electricity generation components. A full report will be published later this year. The analysis and preliminary results are summarized below.

Table 3. Hybrid Electric Vehicle Development Activities

Market	Company /Model	Fuel and power system	Status
Passenger Vehicle	Toyota Prius	Gasoline ICE HEV	Began sales in Japan Dec 10, 1997, has delivered 700 and has 3700 on back order. Monthly production of this car has gone from 1000 to 2000 per month to meet demand. The car travels 66 miles per gallon
	GM Modified EV1	Diesel ICE, Natural gas ICE, and PEM HEV	Experimenting with hybrid electric power systems in the EV1 platform
	Chrysler Intrepid ESX	Diesel ICE HEV	Built a series hybrid electric four door car in 1996, working on a parallel hybrid electric.
	Mitsubishi Space Wagon/ Chariot	Diesel ICE HEV	Mitsubishi is expected to introduce these two cars over the next two years. They have also developed an all-electric Miata.
Transit & School Bus	Bluebird Bus	Diesel, natural gas and H2 ICE HEV	Involved in the hydrogen bus project. Have experimented with natural gas and diesel hybrid electric systems as range extenders for their electric school bus.
	Lockheed-Orion	Natural Gas ICE HEV	Recently won a competitive award to supply 4 diesel hybrid electric buses to New Jersey Transit and 10 diesel hybrid electric buses to New York Transit Authority. Units will be delivered in 1998. Future plans will depend on the performance of these units in the field.
	APS Systems	Prpoane ICE HEV	Plan to put one prototype bus into service in Calfornia in 1998. Nickel cadmium batteries, propane rotary engine.
	Advanced Vehicle Systems	Natural Gas ICE HEV	Built two natural gas hybrid electric buses to be used in Chattanooga, Tennessee.
Military	TDM	Diesel ICE HEV	Won an award to build 10-15 hybrid electric service vehilcles for the US Army.
	Unique mobility HE hummer	Diesel ICE HEV	Developed a hybrid electric hummer for the U.S. Army. Offers improved performance over conventionally-powered hummer. Also, can run in "Stealth mode," running only on the electric motor.

Table 4 shows the curb weights of several diesel, natural gas, and hydrogen-fueled transit buses. Notice that the curb weight of the diesel hybrid electric vehicle is 4,430 pounds higher than the conventional diesel ICE, mostly due to the weight of the batteries. The gross vehicle weight for a diesel hybrid is increased to 38,000 pounds giving a net reduction in people-carrying capacity of 1,430 pounds, or nearly 20%. The natural gas and hydrogen options are heavier still because of the increase in on-board fuel storage and ICE weight compared to diesel. This incremental weight directly displaces people-carrying capacity because the two-axle bus gross vehicle weight is limited to 38,000 pounds by the weight-bearing capacity of the roads.

The data in Table 4 indicate that reductions in the weights of the batteries and power systems are important to developing hybrid electric buses. In reduced range applications or smaller size transit buses the weight limitations may not be as severe.

Table 4. Weights of Conventional and HEV buses

	Diesel		Natural Gas	Hydrogen	
Bus Component	ICE	Hybrid	Hybrid	Pressure Vessel	Mg Hydride
Batteries	0	4,000	4,000	4,000	4,000
Fuel Storage[1]	200	130	170	650	1,900
ICE[2]	2,000	1,400	2,100	2,800	2,800
Motor/ Generator[3]	0	600	600	600	600
Balance	24,800	25,300	25,300	25,300	25,300
Vehicle Curb Weight	27,000	31,430	32,170	33,350	34,600
Gross vehicle weight limitation	36,000	38,000	38,000	38,000	38,000
Passenger weight capacity	**8,000**	**6,570**	**5,830**	**4,650**	**3,400**

[1] Based on a 200 mile range
[2] Diesel engine weight based on 275 hp Navistar Engine; 30% power reduction for HEV; multipliers of 1.5 and 2.0 used for natural gas and hydrogen ICE's respectively
[3] Estimates of advanced systems from Solectria and Fisher Electric Technology

A transit bus will be driven anywhere from 20,000 to 40,000 miles per year, and fuel cost is an important part of the overall bus cost. Figure 4 shows the fuel cost for diesel, natural gas, and hydrogen buses over a range of hydrogen fuel costs. A first-of-a-kind natural gas reformer sized to fuel 40 buses could deliver compressed hydrogen at a cost of 5 $/kg (Thomas, C.E., et al, 1997). Clearly, low-cost hydrogen is needed to enable commercialization of hydrogen fueled buses.

The success of the hybrid electric platform hinges largely on the development of a peaking battery. If an acceptable peaking battery is developed and the hybrid electric platform proves commercially viable, on-board natural gas combustion is a strong competitor to hydrogen. Diesel emits relatively large amounts of nitrous oxides and particulate matter and not readily comparable to hydrogen. Natural gas exhibits full life-cycle pollutant and greenhouse gas emissions comparable to hydrogen. It is less expensive than hydrogen on a delivered-energy basis, and the efficiency differential between a hydrogen-fed PEM fuel cell and a natural gas ICE are reduced in a hybrid electric platform. (This is driven by the fact that the level operation of the ICE in a hybrid electric platform greatly increases its efficiency, and the parallel hybrid, a new development in hybrid electric vehicles that offers increased fuel efficiency, is not amenable to a fuel cell. A parallel hybrid is a hybrid of a hybrid in which mechanical torque from the ICE can be transferred directly to the wheels when it is optimal to do so. Since a fuel cell does not generate torque, it cannot utilize a parallel hybrid configuration.)

The question then is how can a hydrogen bus be differentiated from a natural gas bus to become preferable? One possible differentiating characteristic is carbon dioxide emissions and global climate change. Because a vehicle with on-board hydrogen storage is carbon-free, it is potentially carbon dioxide emission-free. However, the development of renewable-based hydrogen production technologies and/or carbon dioxide sequestration systems are required to realize zero full life-cycle carbon dioxide emissions. Another potential motivation characteristic is robustness of pollutant emissions. Natural gas ICE systems rely on tailpipe clean-up to achieve low emissions and the performance of the clean-up device may degrade over time and during transient conditions.

Advanced hydrogen storage options being developed by the Hydrogen Program can also differentiate hydrogen from natural gas. These are low-pressure systems that do not require a high-pressure gas compressor and have markedly different vehicle safety characteristics. Hydride and fullerene near- to mid-term options require heat at 120 to 250 °C to release hydrogen and so require an on-board heat balance. This is difficult to achieve with a PEM fuel cell that rejects heat at 60-80 °C. Internal combustion engines operate at 300-400 °C and are a better heat match with advanced hydrogen storage options.

Energetics hopes to pursue a more rigorous analysis of the on-board heat balance issue associated with advanced hydrogen storage options, and assess design tradeoffs among parasitic fuel loss, heat exchanger size, and power system efficiency and operating temperature.

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Figure 1. Schematic of the HBr-based Electricity Storage System

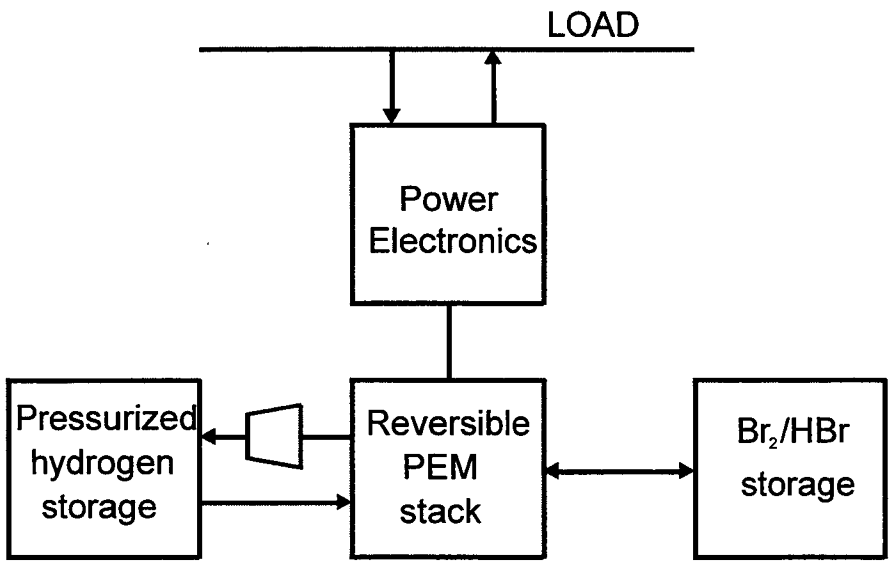


Figure 2. Financial Analysis of a 300 kW, 8 hour HBr-based Electricity Storage System

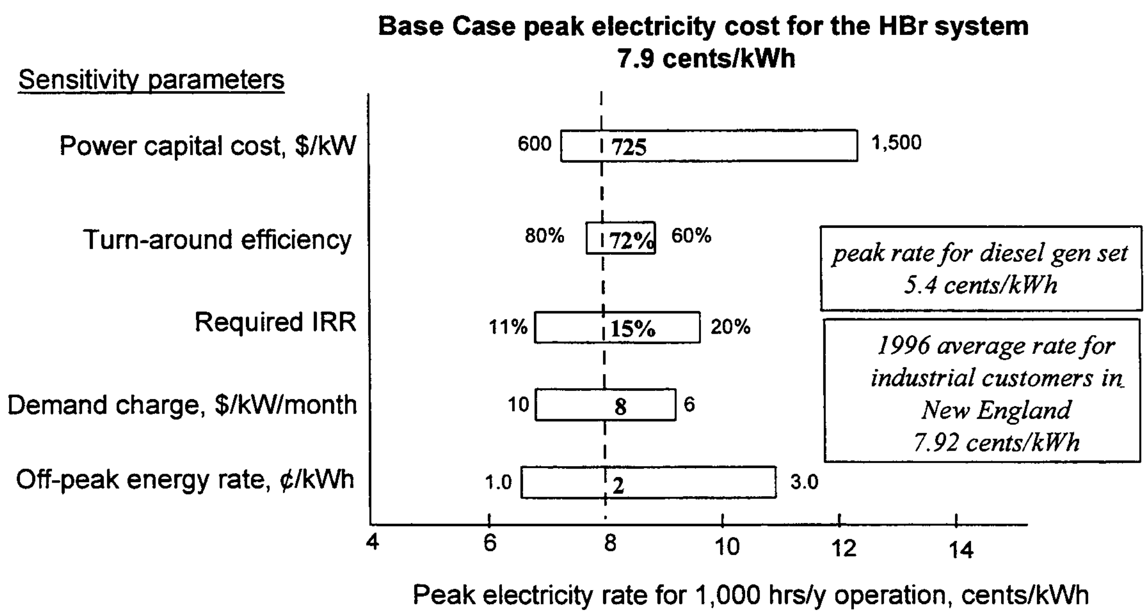


Figure 3 Weight and Volume Density of Hydrogen Storage Technologies

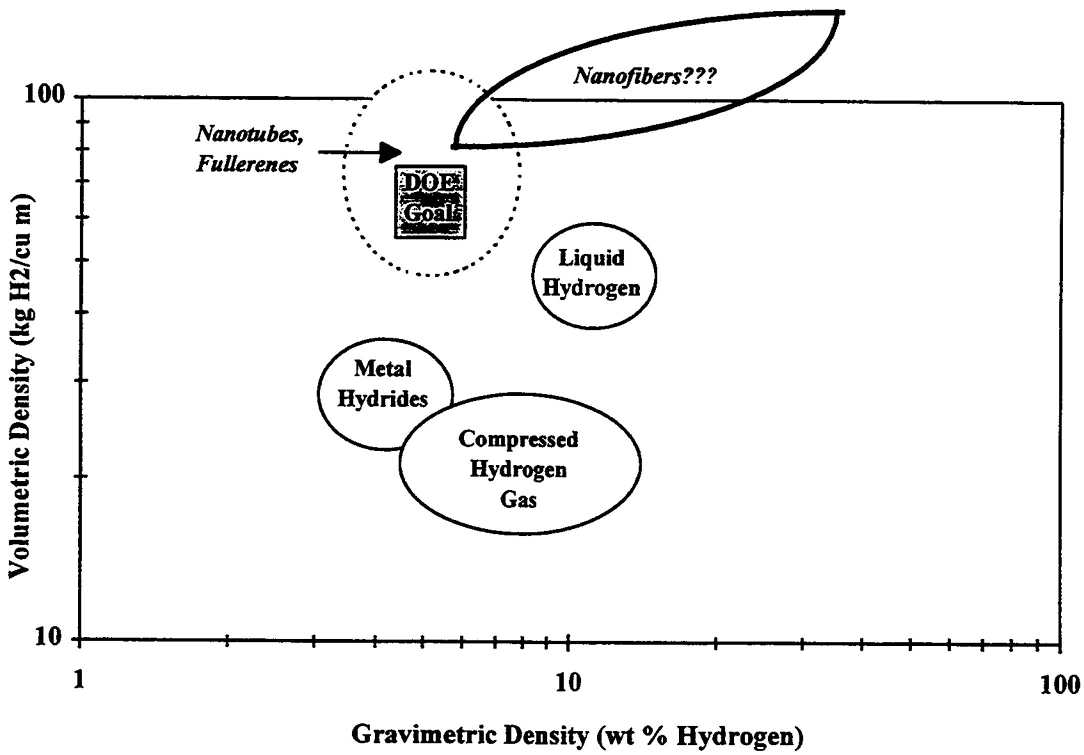
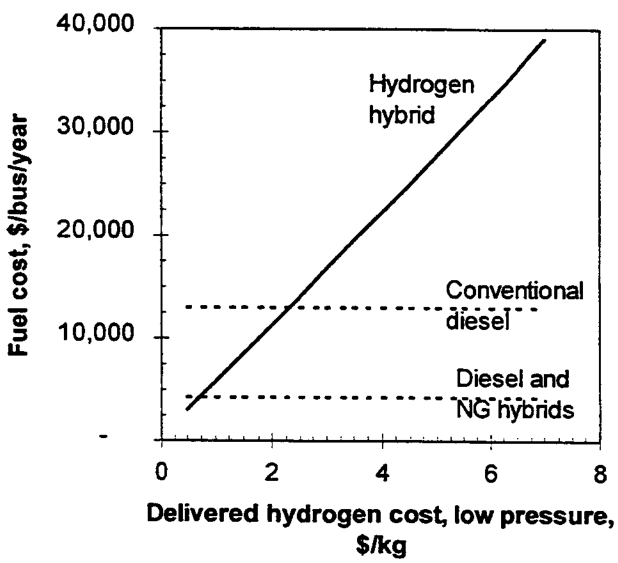


Figure 4. Fuel Cost for Transit Buses



Based on 40,000 miles per year, diesel cost 1 $/gallon,
NG cost 2 cents/kWh, conventional diesel eff 3 mpg