Hydrogen Fuel Cell Vehicle Study

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INTRODUCTION

At the beginning of 2002, the Bush Administration announced the "FreedomCAR" initiative, an industry-government cooperative effort, to develop fuel cell vehicles. This prompted a subcommittee of the POPA Energy and Environment Committee to commence work on a report about fuel cells and FreedomCAR. The rationale for preparing such a report is that the topic is an important aspect of the nation's energy policy—a topic that physicists justifiably feel competent to discuss. Previous POPA studies have been on nuclear energy, energy supplies, etc. Fuel cells are of interest to the physics community (*e. g.,* see the recent *Physics Today* article by Joan Ogden [1]) and physicists are actively involved in research areas for potential hydrogen storage, such as carbon nanotubes. The materials aspects of fuel cells are especially within the purview of physicists. Overall systems considerations, wells-to-wheels energy efficiency, and related issues can benefit from analysis by physicists. In view of the high expectations for fuel-cell vehicles generated by the FreedomCAR initiative, it seems reasonable to examine what is reality and what is unsupported optimism. Of those who have read the Ogden article or popular-press fuel cell articles, some will want to know more. This report is a start on a balanced discussion that intends to educate, rather than persuade or advocate. The intended audience is POPA and the APS membership. [See Appendix A for fuel cell principles of operation and a schematic.]

The motivation for the FreedomCar initiative is to reduce U.S. dependence on imported petroleum, to reduce emissions of atmospheric pollutants, and to reduce CO_2 emissions by improving fuel economy and/or by going to a hydrogen-based system. Since the transportation sector itself uses more oil than produced domestically (Fig. 1), FreedomCAR also addresses a serious national security issue.

The big three automotive manufacturers have publicly committed their companies to participation in the initiative. General Motors Chairman Jack Smith: "With the FreedomCAR program, we are taking a major step towards creating a future where the vehicle is no longer part of the energy and environmental debate." DaimlerChrysler CEO Dieter Zetsche: "FreedomCAR focuses on jointly developing technologies that are important to the entire automotive industry. This program allows us to continue to work together as an industry in a way that can make a difference." Ford Chairman and Chief Executive Officer William Clay Ford Jr.: "Our companies have made significant progress in reducing the environmental impact of our products. Our participation in FreedomCAR signifies our commitment to continue that progress."

FreedomCAR has the following technology-specific goals for 2010 [2] [See Appendix B for a complete set of goals and notes.]:

- To ensure reliable systems with costs comparable with conventional internal combustion engine/automatic transmission systems, future fuel cell powertrains should have
    - Electric propulsion system with a 15-year life capable of delivering at least 55 kW for 18 seconds and 30 kW in a continuous mode, at a system cost of $12/kW peak. [Note this pertains to electrical systems other than the fuel cell such as electric motors, controllers, etc.]

- A durable fuel cell power system (including hydrogen storage) that achieves 60% energy efficiency when operating at peak power and that offers a 325 W/kg power density and 220 W/L operating on hydrogen. Cost targets are $45/kW by 2010, $30/kW by 2015.

- To enable clean, energy-efficient vehicles operating on clean, hydrocarbon-based fuels powered by fuel cells, the goal is
    - Fuel cell systems, including a fuel reformer, that have a peak energy efficiency of 45% and meet or exceed emissions standards with a cost target of $45/kW by 2010 and $30/kW in 2015.

- To enable the transition to a hydrogen economy, ensure widespread availability of hydrogen fuels, and retain the functional characteristics of current vehicles, the goals are
    - Demonstrated hydrogen refueling with developed commercial codes and standards and diverse renewable[1] and non-renewable energy sources. Targets: 70% energy efficiency well-to-pump; cost of energy from hydrogen equivalent to gasoline at market price, assumed to be $1.25 per gallon (2001 dollars).
    - Hydrogen storage systems demonstrating an available capacity of 6 wt% hydrogen, specific energy of 2000 W-h/kg [pertains to storage system mass], and energy density of 1100 W-h/L at a cost of $5/kWh.

- To improve the manufacturing base, the goal is
    - Material and manufacturing technologies for high-volume production vehicles that enable and support the simultaneous attainment of
        - 50% reduction in the weight of vehicle structure and subsystems,
        - affordability, and
        - increased use of recyclable/renewable materials.

An expanded initiative, entitled the FreedomCAR and FUEL initiative, that focuses more on infrastructure issues, has been announced recently.

> "Tonight I am proposing $1.2 billion in research funding so that America can lead the world in developing clean, hydrogen-powered automobiles."
>
> — President George W. Bush, State of the Union Address, January 28, 2003

Technical goals are not yet available, but the general goals are [2]:

- **Lowering the cost of hydrogen:** Currently, hydrogen is four times as expensive to produce as gasoline (when produced from its most affordable source, natural gas). The FreedomCAR and Fuel Initiative seeks to lower that cost enough to make fuel cell cars cost-competitive with conventional gasoline-powered vehicles by 2010; and to advance the methods of producing hydrogen from renewable resources, nuclear energy, and even coal.
- **Creating effective hydrogen storage:** Current hydrogen storage systems are inadequate for use in the wide range of vehicles that consumers demand.
- **Creating affordable hydrogen fuel cells:** Currently, fuel cells are ten times more expensive than internal combustion engines. The FreedomCAR and FUEL Initiative is working to reduce the cost to affordable levels.

[1] "Renewable" energy generally refers to solar, wind, geothermal, tidal, hydroelectric energy, and biomass.

Not everyone was pleased. In the February 2, 2003 issue of the *New York Times*, several groups (Cato Institute, National Taxpayers Union, the Competitive Enterprise Institute, and the Natural Resources Defense Council) listed the "hydrogen car" as a prominent item to cut from the federal budget to reduce the projected shortfall.

GLOSSARY

ICE	Internal combustion engine
NG	Natural gas
CNG	Compressed natural gas
SI	Spark ignited
HEV	Hybrid electric vehicle
IC	Internal combustion
LHV	Lower heating value
CI	Compression ignited
SIDI	Spark ignited direct injection
CIDI	Compression ignited direct injection
FC	Fuel cell
GNF	Graphite nanofiber
SWNT	Single-walled carbon nanotube
MWNT, CNT	Multi-walled carbon nanotube

SYSTEM CONSIDERATIONS

In the April, 2002 issue of *Physics Today* Joan Ogden of the Princeton Environmental Institute discussed the future of hydrogen as a fuel and described the operation of hydrogen-oxygen fuel cells. Ogden stated that practical fuel cells[2] are up to 60%

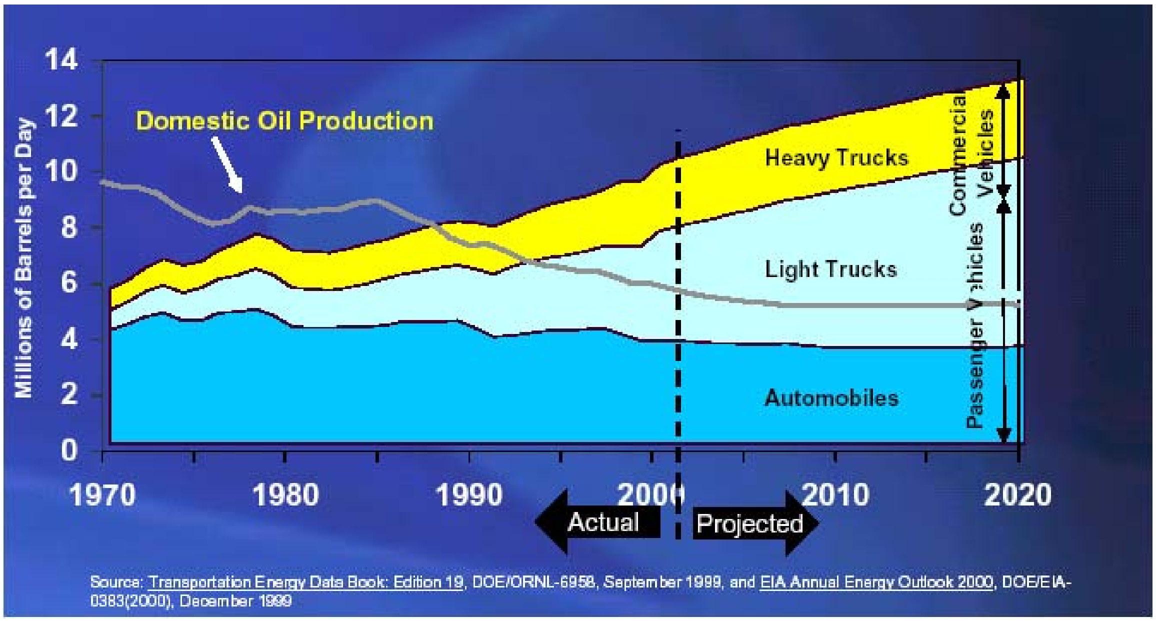


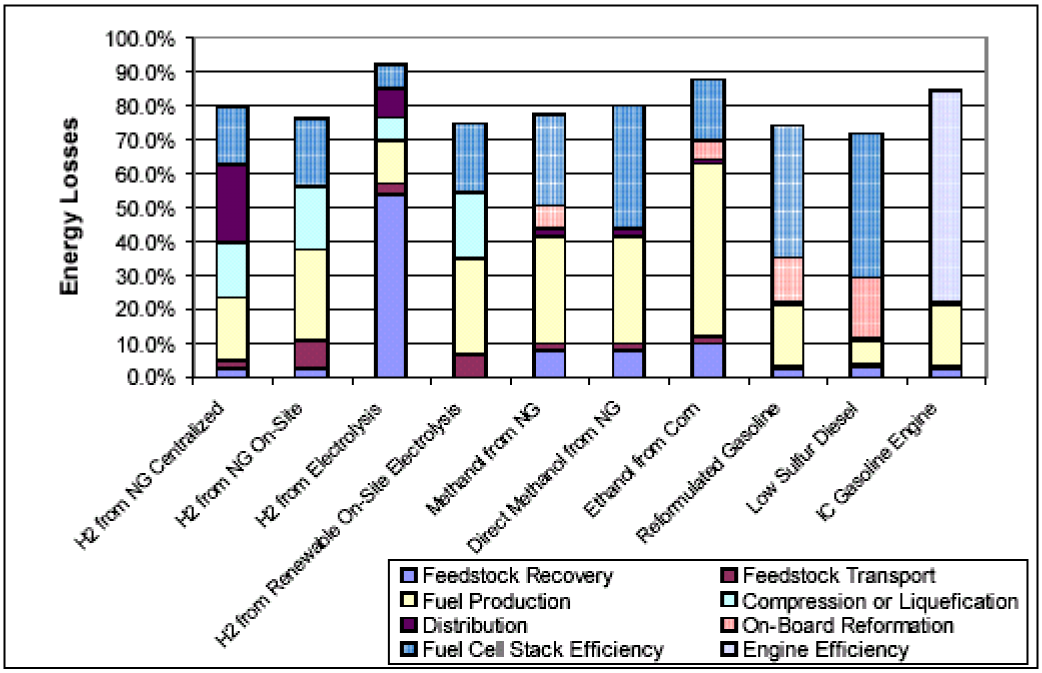
Fig. 1 Taken from Ref. [3].

efficient in converting hydrogen energy into electrical energy (which is the FreedomCAR target) although not necessarily at the rated power, significantly higher than the 45% efficiency of using hydrogen in an internal combustion engine. However, these estimates do not include the losses in producing hydrogen from various hydrocarbon sources (Fig. 2). Clearly, hydrogen is not a naturally occurring terrestrial fuel. Rather, it is an energy carrier.

A typical well-to-wheels analysis is shown in Fig. 3. Although the details of the analyses behind Fig. 2 are not readily available, the kind of breakdown of energy losses shown in Fig. 3 underlies each the powertrain options considered.

[2] We refer to proton-exchange membrane fuel cells in this report.

Net Energy Losses "Wells to Wheels" for Fuel cell Vehicles



Sources: Albert Sorbey and Associates, 1996; Pembina Institute for Appropriate Development, 2000; Karlhammer, 1997; Moore, 2000; Thomas et al., 1998; and Stodolsky et al., 1999.

Fig. 2 Due to way this graph has been drawn, to obtain the energy efficiency of any system, subtract the height of the bar from 100%. From Ref. [4].

For example, the equivalent fuel economy of a compressed natural gas, spark ignited, hybrid electric vehicle (CNG SI/HEV) is 48.6 mpg, whereas a fuel cell vehicle powered by hydrogen derived from methane is projected to get 82.0 mpg, a substantial improvement. However, if viewed from the standpoint of well-to-wheels energy consumed per unit of distance traveled, the difference is more modest: 2867 versus 2368 BTU/mi. If CO_2 is sequestered in the forming of hydrogen, the amount emitted into the atmosphere is only 25 g/mi for the fuel cell vehicle compared to 196 g/mi for the CNG vehicle. This additional benefit favors the fuel cell vehicle. Clearly hydrogen fuel cells do not entirely eliminate CO_2 emissions unless the hydrogen is generated without combusting or reforming hydrocarbon fuels, *e.g.* by electrolysis of water using nuclear, solar or wind power. The emission of CO, NO_x and hydrocarbons associated with the ICE are removed, but may be emitted to some extent in a different location by the chemical plant generating H_2.

Figure 9-3 "WELL-TO-WHEEL" EFFICIENCY FOR VARIOUS VEHICLE SCENARIOS

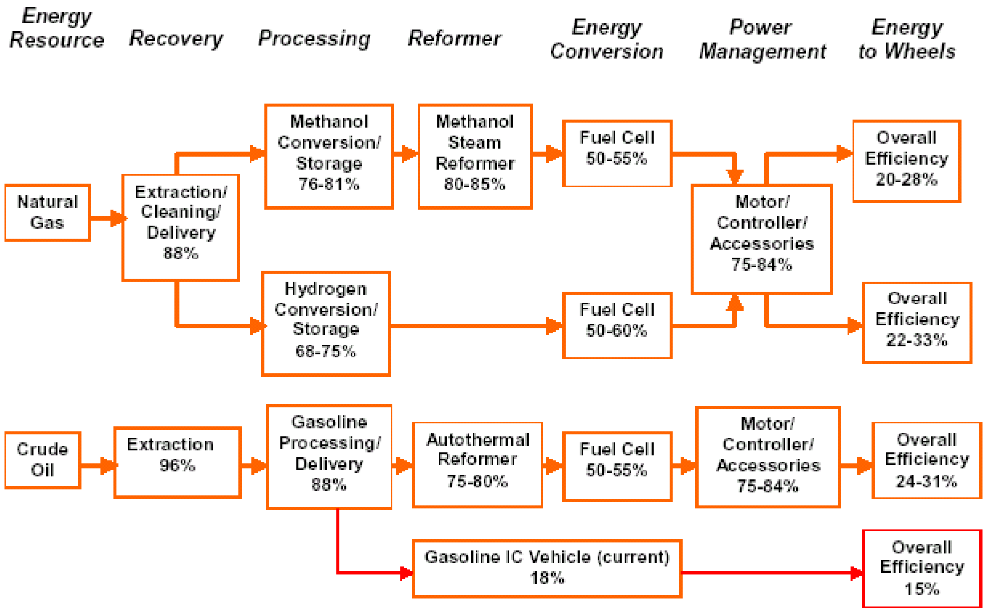


Fig. 3 From Ref. [5].

Even if production losses are taken into account, the fuel cell vehicle surpasses the conventional internal combustion engine in efficiency, although the overall efficiency is only about 30% in the best case, less than the 60% x 70% = 42% well-to-wheels efficiency objective of FreedomCAR (Fig. 4), where 60% is the energy-to-wheels goal and 70% is the well-to-pump efficiency.

Further comparison of internal combustion engine and fuel cells for fuel economy and CO_2 emissions is shown in Table I.

In a draft report prepared last year for the Office of Transportation Technologies, Energy Efficiency and Renewable Energy, U.S Department of Energy, [6] national lab scientists and others indicated an optimistic mid-term future for fuel cell vehicles (Table II).

Ogden pointed out major obstacles that must be overcome before automotive fuel cell technology can be considered viable. First, today's cost of $1500 to $10 000 per kilowatt of power must come down to the range of $50-100 per kilowatt to be competitive. According to Ogden, the most expensive component is the membrane electrolyte, typically made of the polymer Nafion. Also, A.D. Little has indicated that the current platinum requirement for a 50-kW system would cost $57/kW, which is higher than the FreedomCAR cost target for the entire fuel cell system. [7][3] [See Appendix C for another cost analysis.] Second, a breakthrough in on-board hydrogen storage is required. The currently preferred method is to use a carbon-fiber wrapped compressed-gas cylinder

[3] This analysis, however, seems high. By 2000, Pt usage had been reduced to about 0.5 g/kW, [8] which gives a cost of roughly $10/kW for the catalyst.

Table I. Fuel Economy, Energy Use and CO_2 Emissions for Alternative Fueled Automobiles [9].

	FUEL ECONOMY Mpg equiv- LHV basis (from GREET model; except fuel cell vehicles and H_2 ICE/HEVs from DTI)	Well to Wheels Energy Consumption (BTU/mi)	Well to Wheels CO_2 emissions (g/mile)
IC ENGINE VEHICLES			
Conventional Gasoline SI Engine	*22.4*	6492	514
CNG SI Engine	*20.3*	6702	459
Adv. Diesel CI Engine	*37.0*	4565	378
ICE/HYBRID VEHICLES			
Gasoline SIDI/HEV	*46.9*	3092	252
CNG SI/HEV	48.6	2867	196
Ethanol SIDI/HEV	*46.9*	4921	67
H_2 SI/HEV	**50.0**	3466 w/o CO_2 seq 3580 w/CO_2 seq	234 w/o CO_2 seq 41 w/ CO_2 seq
Diesel CIDI/HEV	*56.8*	2487	208
FUEL CELL VEHICLES			
Gasoline (probable) (best)	**38.0** **49.4**	3819 2938	304 234
Methanol (probable) (best)	**56.0** **64.2**	3212 2802	199 174
Hydrogen (from natural gas with steam reforming, pipeline delivery and compression to 5000 psi for onboard storage	**82.0**	2368 w/o CO_2 seq 2446 w/CO_2 seq	143 w/o CO_2 seq 25 w/CO_2 seq.

Fuel economies shown in *italics* are from the GREET model [10]. Fuel economies in boldface are from the DTI model [11]. CNG SI/HEV vehicle fuel economies have been scaled from GREET results.

Table II. Source: Ref. [6].

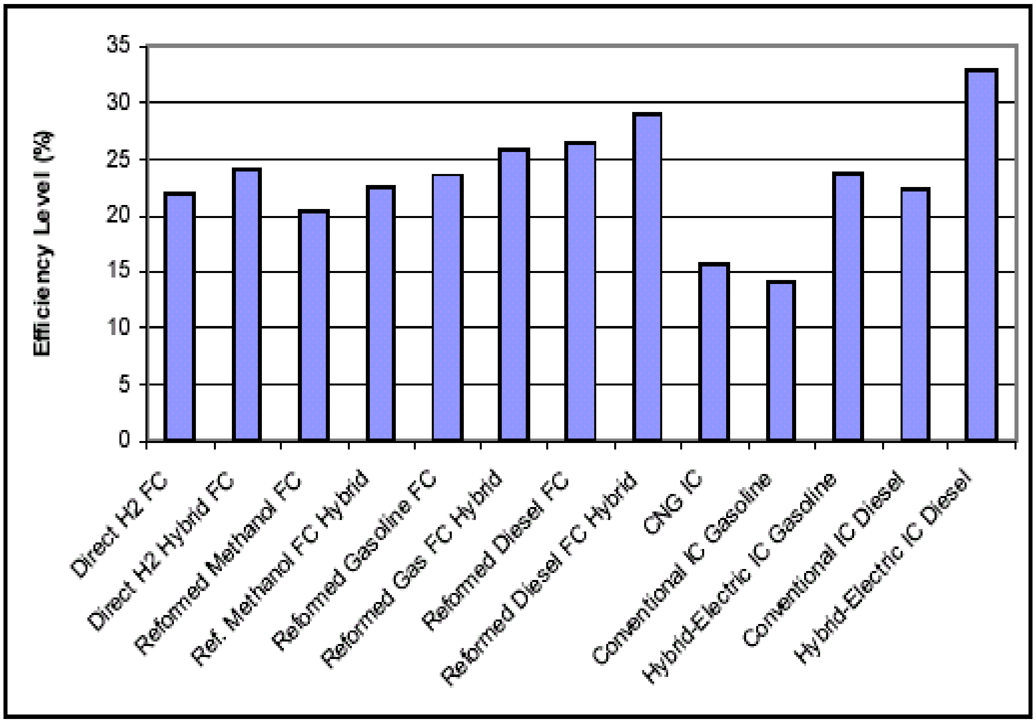
Vehicle System	Fuel Economy Improvement Potential	Criteria Emissions	Years to Mass Market Introduction	Current Incremental Cost	Other Issues
Enhanced Conventional	Moderate (50%)	Continued though reduced	Very near term (0-5 y)	Minimal (5%)	High consumer acceptance, continued petroleum dependence
Hybrid	Substantial (100-200%)	Some zero emission range possible	Near term (2-7 y)	Substantial (10-20%)	Grade climbing ability or towing capacity may be reduced
Fuel Cell	Very High (150-300%)	Low to zero tailpipe and total	Mid term (7-12 y)	Very high (>20%)	Potential petroleum independence
Battery-Electric	Very High (300%)	Zero tailpipe	Near term (2-7 y)	Very high (>20%)	Energy storage, range concerns, low petroleum use

(at a pressure of 34 MPA or 5000 psi, with mass of 32.5 kg, and volume of 186 L for a 500-km range). [See the next section for a discussion of hydrogen storage technology.] An infrastructure to produce and distribute hydrogen economically is the third major problem to be solved. Presently most hydrogen is produced thermochemically (500-1700 C) in oil refineries and chemical plants by reforming natural gas and other hydrocarbons with steam or oxygen. Unlike petroleum, natural gas supplies are abundant and come mostly from within the United States or are imported from Canada. Production facilities operate at approximately 70% of capacity and the distribution infrastructure has excess capacity [4]. Thus, at least initially, natural gas production and distribution does not appear to be a limiting factor in the availability of hydrogen.

A different approach that does not rely on hydrocarbons has been analyzed by C. W. Forsberg and K. L. Peddicord [12]. They discussed the economics of H_2 production using nuclear energy to provide the energy for electrolysis of water and concluded "The technology has the potential for economic production of H_2." Likewise, hydrogen production from renewable sources such as wind power could be interesting, but has not been analyzed here. Fortunately, the distribution of H_2 may not be as daunting as one might think. Fosberg, in a private communication, noted the existence of several hydrogen pipelines in Europe, the United States and Japan. However, natural gas lines would have to be retrofitted with new valves and compressors before hydrogen could be transported through them.

Initially the auto industry felt that sufficient hydrogen fuel would not be available quickly enough, so engineers pursued a path that required on-board reforming of gasoline or methanol. DaimlerChrysler demonstrated an example in October 2000. The Jeep Commander 2 (similar to the Jeep Grand Cherokee sport utility vehicle) reformed pure,

Comparison of Energy Efficiencies for Fuel Cells, Internal Combustion and Hybrid Vehicles



Sources: Stodolsky et al., 1999; Albert Sorbey and Associates, 1996; Pembina Institute for Appropriate Development, 2000; and Thomas et al., 1998.

Fig. 4 From Ref [4].

electronic-grade methanol to power two Ballard fuel stacks. Although DaimlerChrysler demonstrated 23.5 mpg fuel efficiency (almost twice that of a comparable gasoline vehicle) with acceptable performance and acceleration, they found that fuel reforming must be improved because the cold-start time was unacceptable [13].

On May 1, 2002, according to a press release, General Motors demonstrated the world's first drivable fuel cell vehicle (a Chevrolet S-10 fuel cell pickup) that extracts hydrogen from gasoline. "This vehicle and the reforming technology in it move us closer to a hydrogen economy," said Larry Burns, GM's Vice President of Research and Development, and Planning. The fuel cell pickup was equipped with a fuel processor that reformed low-sulfur gasoline. When linked with a fuel cell stack, GM said the vehicle could achieve up to 40 percent overall energy efficiency, which is a 50 percent improvement over a conventional internal combustion engine. For further information, see Burns *et al.* [14].

Ron Sims, Ford Motor Co. research engineer (retired) and consultant to ORNL, feels that gasoline reformers (and presumably methanol as well) on-board the vehicle are no longer viable because they are too costly and too complex. Obviously, the reformer adds another chemical plant to the vehicle—an undesirable feature. However, stationary reformers at gas stations might make sense.

Sims thinks it will take 10-15 y for commercialization of fuel cell vehicles, 20 y before internal combustion engine sales will notice the impact of fuel cells. On the other hand, Larry Burns and other GM executives have publicly stated, “By the end of this decade, you can expect to see affordable, profitable fuel cell vehicles on the road."

By 2000, Ford had built a hydrogen refueling station at the Engineering and Research Center in Dearborn, Michigan (Fig. 5) and had developed a hydrogen (no reforming) fuel cell vehicle with on-board storage of compressed gas (Fig. 6).

According to Ref. [14] Toyota is preparing a fuel cell hybrid vehicle, called FCHV-4, for production. Two vehicles have been delivered to the University of California for research purposes [15]. The Toyota vehicle uses compressed hydrogen gas, as does the Honda FCX, currently being tested in California. The 2003 Honda FCX has just been certified in the US as a zero emission vehicle. [16] General Motors exhibited a new prototype fuel cell hybrid named “Hy-wire” at the Paris Motor Show in September 2002. Although this vehicle has a top speed of 100 mph, it has a range of only 100 miles, far short of the acceptable driving range of 300 miles. In the May 2002 press release Larry Burns, while still maintaining that GM will produce affordable, customer-friendly fuel cell vehicles by 2010, believes GM will only “sell them <u>profitably and in large numbers</u> by 2020.” Clearly the timetable is rather long, consistent with the opinion of Ron Sims. For more information on prototype fuel cell vehicles, see the article by Jost [17] and Ref. [18].

Other engineering issues that all manufacturers face, although seemly mundane, are nonetheless challenging:

- Cold weather operation
- Packaging
- Reliability
- Safety (including public acceptance of H_2 fuel) [See section on safety below.]
- Manufacturability

In an interview we had with Prof. J. Schwank, U. of Michigan Chemical Engineering Dept., he suggested that the hydrogen internal combustion engine is potentially a better bet than fuel cells because hydrogen does not then have to be as highly purified. The ICE can take as much as 10% CO in the fuel, which would be lethal for the fuel cell. Purification of the hydrogen stream to remove CO is a significant cost. In Table I, the fuel economy of the H_2 SI/HEV (spark-ignited, hybrid-electric vehicle) is estimated to be 50 mpg with about 3500 BTU/mi (2.3 MJ/km) well-to-wheels energy consumption compared to 2400 BTU/mi (1.6 MJ/km) for fuel cells.

Schwank still feels that fuel processors, which reform hydrocarbon fuels to produce hydrogen, are important and they need to be highly flexible because of the difference in

Fig. 5 Source: Ref. [8].

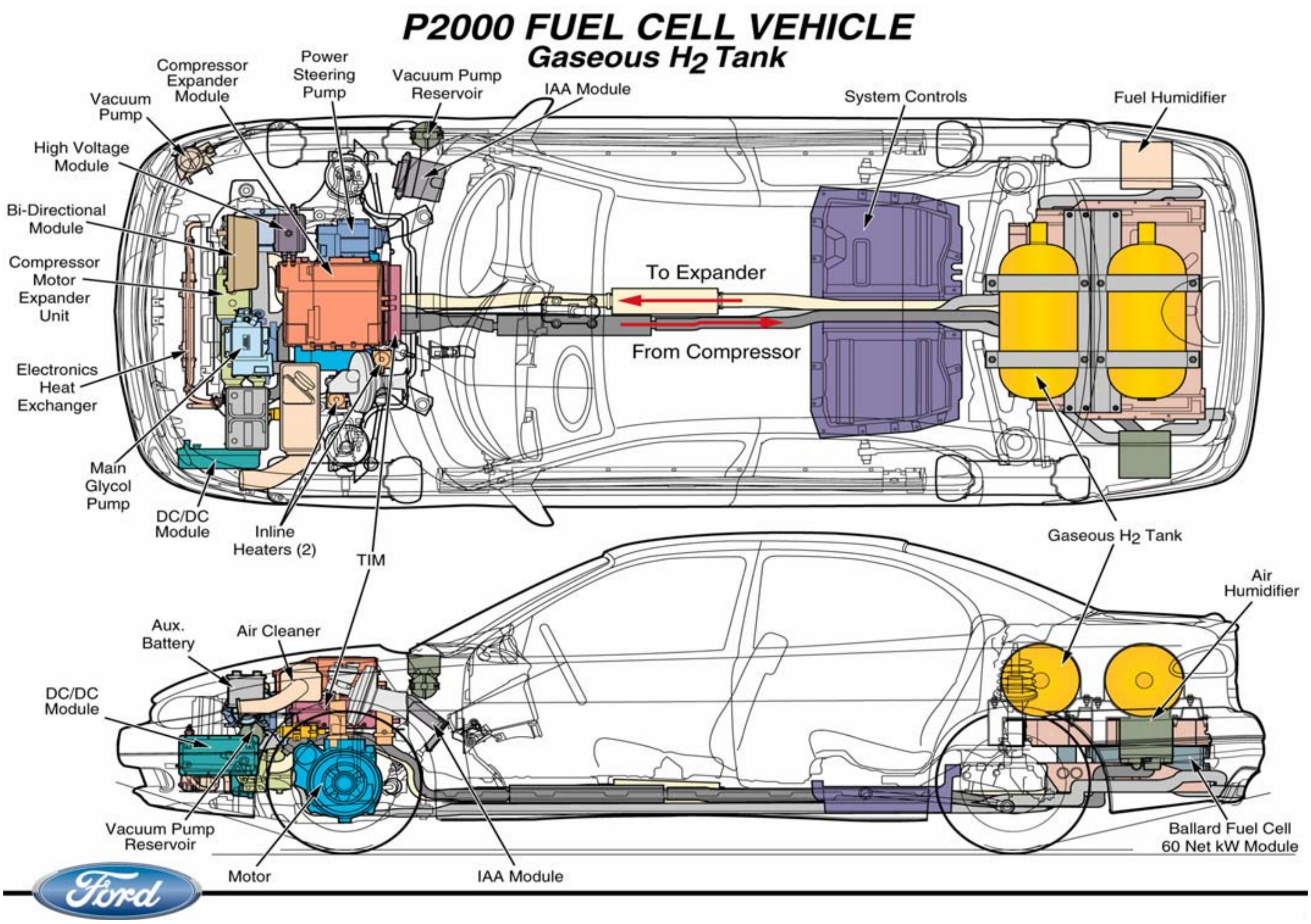


Fig. 6 Source: Ref. [8].

fuels by season and region of the country. For military applications, the processor must be able to run on the highly developed (and unlikely to be changed!) JP8 fuel.

The Army estimates it costs $600/gal to carry fuel to the battlefield, so reducing consumption is essential. Instead of powering vehicles, fuel cells could be used as auxiliary units to run on-board electronics and communications equipment. Quiet operation to generate electricity for stationary field applications is just as important to the military as higher fuel efficiency. The Army is presently letting contracts for demonstration fuel cell powered trucks [19]. The impact that military funding might have for fuel cell development is huge.

In addition to USCAR, which is the automotive industry consortium that has responsibility for implementing many of the FreedomCAR and FUEL programs, the California Fuel Cell Partnership [20] is actively testing, promoting, and demonstrating fuel cell technology. This partnership of automotive manufacturers, energy companies, fuel cell companies, and government intends to evaluate as many as 60 vehicles in real-world conditions.

Recently, MIT issued a detailed report comparing life-cycle energy costs that includes energy expended in manufacturing and final disposal as well as for fuel consumption on the road and during production and distribution. Weiss *et al.* [21] made reasonable assumptions about the future evolution of internal combustion engines and hybrid-electric vehicles in addition to predicting the pace of development for fuel cell vehicles. On the basis of life-cycle energy consumption, the differences between fuel cell and hybrid-electric vehicles (mid-size) are small compared to the uncertainties in the predictions. The manufacturing energy requirements do not differ substantially, but the predicted fuel economy of a 2020 diesel ICE hybrid electric is high enough and the fuel cycle energy low enough to offset much of the advantage of fuel cells. [See Appendix D.] Their study pertains to the next twenty years and to energy derived from petroleum or natural gas. Similar conclusions were found for green house gas emissions.

HYDROGEN STORAGE TECHNOLOGIES

Introduction

In the development of fuel cell vehicles, hydrogen storage is "the biggest remaining research problem" according to the January 2003 Office of Technology Policy report, *Fuel Cell Vehicles: Race to a New Automotive Future* [22]. Current hydrogen storage systems are inadequate to meet the needs of consumers in a fuel cell vehicle. The OTP report continues, "Hydrogen's low energy-density makes it difficult to store enough on board a vehicle to achieve sufficient vehicle range without the storage container being too large or too heavy."

Existing and proposed technologies for hydrogen storage include (1) physical storage: pressurized tanks for gaseous hydrogen and pressurized cryotanks for liquid hydrogen; (2) reversible hydrogen uptake in various metal-based compounds including hydrides, nitrides, and imides; (3) chemical storage in irreversible hydrogen carriers such as methanol; (4) cryoadsorption with activated carbon as the most common adsorbent; and (5) advanced carbon materials absorption, including carbon nanotubes, alkali-doped carbon nanotubes, and graphite nanofibers. The U.S. Department of Energy report, *A National Vision for America's Transition to a Hydrogen Economy—To 2030 and Beyond*, projects that pressurized tanks will be the predominant hydrogen storage technology until about 2015, to be supplanted by hydride storage into the early 2020s, then other solid state storage technologies [23]. They see storage technologies maturing sufficiently for mass production in the 2020s.

The Department of Energy timeline for development of storage systems projects that high pressure and cryogenic storage will be demonstrated in 2002-3, cost-effective hydride storage systems in 2003-6, and carbon-based storage systems in 2006-11 [24].

Goals for hydrogen storage systems for 2010 that were established in the FreedomCAR initiative [2] include

- available capacity of 6 wt% hydrogen
- specific energy of 2 kWh/kg
- energy density of 1.1 kWh/L
- cost ≤ $5/kWh or $1.25/gal (gas equiv.) in CY2001 dollars

Research into hydrogen storage technologies is still in its infancy, as reflected in the very low level of patenting in this area: 14 patents in 2001, and fewer in most previous recent years [25]. Patent data indicate that the United States is the "leader" in this research area, while "US-based Energy Conversion Devices and Canadian organization Hydro-Quebec and McGill University is where the action appears to be located" [26]. Likewise, only a few papers on hydrogen storage were presented at the recent (2003) March APS Meeting.

According to John M. Decicco [27], as quoted in Ref. [22], p. 24, hydrogen storage faces deployment barriers that are

- fundamental (basic research still needed)
- developmental (engineering R&D required for practical designs)
- maturational (mass-production commitments premature)
- experience-limited (costs higher than long-run potential due to lack of production experience)
- infrastructural (lack of appropriate fuel or service facilities)

This report summarizes the current status of hydrogen storage technologies and directions of current and needed research in this area.

Pressurized Tank Storage

Pressurized tanks of adequate strength, including impact resistance for safety in collisions, have been made of carbon-fiber wrapped cylinders. Compressed gas storage in such tanks has been demonstrated at a pressure of 34 MPa (5,000 psi) with a mass of 32.5 kg and volume of 186 L, sufficient for a 500-km range. Note, however, that this tank volume is about 90% of a 55-gallon drum, rather large for individual automobiles. So while the 6 wt% goal can be achieved, tank volume is problematic. Pressures of 70 MPa (10,000 psi) have been reached, and in 2002 Germany certified Quantum Technology's 10,000 psi on-board storage tank [24]. A footnote in the OTP report cited above [22] says, "The Toyota and Honda vehicles available for lease in late 2002 use hydrogen stored in high-pressure containers [28]. However, their range will be less than optimal because hydrogen's low density does not permit a sufficient amount to be stored (unlike CNG [compressed natural gas], which has a higher energy density for the same volume)."

Low temperature storage of liquid hydrogen does not appear to be suitable for normal vehicle use, although research on this possibility is being conducted at a low level by several automobile manufacturers [24]. Furthermore, "a liquid hydrogen storage system loses up to 1% a day by boiling and up to 30% during filling, as well as requiring insulation to keep the hydrogen at 20 K." [29]

Hydrogen Uptake in Metal-Based Compounds

Metal hydridation can be used to store hydrogen above room temperature and below 3 or 4 MPa. However, the metals introduce too much additional weight for most vehicle uses. They are also expensive [30].

Recent work by P. Chen, et al. has shown that lithium nitride can reversibly take up large amounts of hydrogen [31]. This material takes up hydrogen rapidly in the temperature range 170-210ºC, and achieved 9.3 wt% uptake when the sample was held at 255ºC for 30 minutes. Under high vacuum (10^{-9} MPa, 10^{-5} mbar, or 10^{-5} torr) about two-thirds of the hydrogen was released at temperatures below 200ºC. The remaining third of the stored hydrogen required temperatures above 320ºC for release. The hydrogen was taken up as lithium imide ($LiNH_2$) and lithium hydride (LiH). These researchers suggest that related metal-N-H systems should be investigated to find a hydrogen storage system that works at more practical temperatures and pressures.

Cryoadsorption Hydrogen Storage

While having potential weight and volume advantages, cryoadsorption with activated carbon as adsorbant requires liquid nitrogen temperatures and 2 MPa (300 psi) to hold the physically adsorbed hydrogen [32]. It does not appear to be suitable for vehicle use.

Carbon Nanotube and Related Storage Technologies

The status of hydrogen storage in advanced carbon materials is still unclear. In this subsection, we review briefly the status of carbon nanotube storage, both single-walled and double-walled, and graphite nanofiber stack storage. Other carbon-based storage technologies that have been proposed include alkali-doped graphite, fullerenes, and activated carbon.

High surface area and abundant pore volume in the nanostructured materials make these especially attractive as potential absorption storage materials. Some early work gave tantalizing results for hydrogen storage in carbon nanotubes. Ogden reported various conflicting, some excessively optimistic, results [1]. A query by this subcommittee to Prof. Mildred Dresselhaus of MIT about the achievable wt% (6.5 wt% has been suggested) brought this response [33]:

> 1. It is hard to say what is a reliable estimate of the hydrogen uptake number because of the differences in the reported levels by different groups, presumably doing similar measurements. The reasons for the different results between groups are not understood.
>
> 2. The 6.5% value is not yet achievable in my opinion.
>
> 3. The problem seems to be hard to me, arguing from a theoretical standpoint. However I would not discount the possibility of a breakthrough that might change the situation dramatically. So far it doesn't seem to me that there is yet much available carefully controlled work.

A 2001 review of carbon nanostructure storage research, sponsored by the German Federal Ministry for Education and Research (BMBF), found that follow-up work "has been unable to reproduce any of the high-capacity results." [29] They concluded "In view of today's knowledge, it is unlikely that carbon nanostructures can store the required amount of hydrogen. In any case, this calls into doubt whether carbon nanostructures would have any advantage over high-pressure tank storage."

The German study [29] presented the following summary figure on the capacity of current and future hydrogen storage systems:

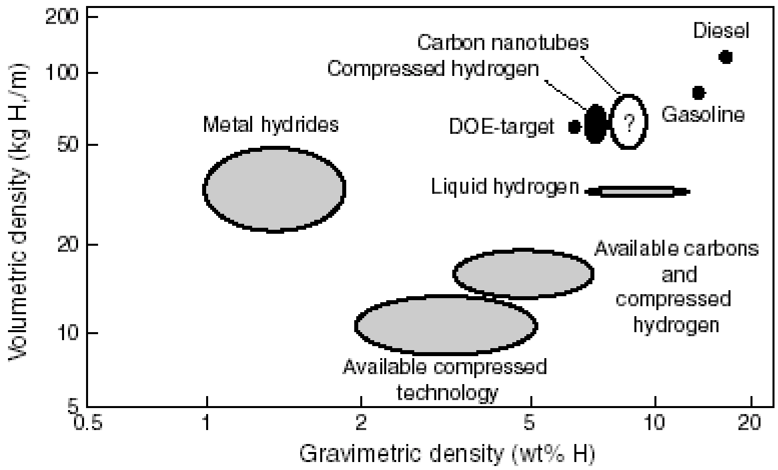


Figure 1. Summary of how current and future hydrogen storage systems relate with respect to gravimetric and volumetric densities.[3]

The considerable doubt that has been cast upon carbon nanotube storage capacity makes it highly likely that their position on this chart will have to be moved to a much less favorable point.

A summary of results was given by Ding et al. [34] and reproduced in Ref. 29.

Among all the reports on carbon nanotube storage capacities, one finds that the reproducible results are invariably lower than the earlier encouraging findings. So values of 1-4 wt% are the best that can be depended upon with present data. These are not sufficient to meet the benchmark of 6 wt%.

A review and research report published in 2002 considered hydrogen storage by graphite, graphite nanofibers (GNFs), and single-walled carbon nanotubes [35]. These authors concluded "These investigations show a reversible hydrogen storage only for SWNTs and in addition indicate that an opening of the SWNTs is essential to reach high storage capacities." Ref. 29 points out that results on GNFs have been mixed and contradictory, with most high capacity results being explained subsequently "by the presence of water vapor, which expanded the spacing between graphite layers (typically ~3.4 Å) to accept multiple layers of hydrogen." [36] Nevertheless, relatively high storage measurements for GNFs (~10-13 wt%) have been reported for GNFs grown by thermal cracking [37] and for vapor-grown GNFs (with some conflicting later results by the same group) [38, 39]. Further research is clearly needed before reliable conclusions can be drawn about GNFs.

Material	Temperature	Pressure, MPa	Max. wt% of H_2	Reference
SWNT 100%	133 K (–140°C)	0.04	5–10 (prediction)	**Dillon 1997**
SWNT High purity	Ambient	0.067	~3.5–4.5	**Dillon 1999**
SWNT ~50%	300 K (27°C)	10.1	~4.2	**Liu 1999**
SWNT High purity	80 K (–193°C)	~7	8.25	**Ye 1999**
MWNT	~300–700 K (27–427°C)	Ambient	0.25	**Wu 2000**
SWNT-$TiAl_{0.1}V_{0.04}$	Ambient	0.067	~7	**Dillon 2000**
SWNT-Ti-6Al-4V	Ambient	0.08	1.47	**Hirscher 2001**
SWNT-Fe	Ambient	0.08	<0.005	**Hirscher 2001**
SWNT ball milled in Ar	Ambient	0.08	<0.1	**Hirscher 2002**
SWNT ball milled in D_2	Ambient	0.9	1.0	**Hirscher 2002**
Li-CNT	473–673 K (200–400°C)	0.1	20	**Chen 1999**
Li-CNT	<313 K (40°C)	0.1	14	**Chen 1999**
Li-CNT (wet H_2)	473–673 K (200–400°C)	0.1	12	**Yang 2000**
Li-CNT (dry H_2)	473–673 K (200–400°C)	0.1	2.5	**Yang 2000**
K-CNT (wet H_2)	<313 K (40°C)	0.1	21	**Yang 2000**
K-CNT (dry H_2)	<313 K (40°C)	0.1	1.8	**Yang 2000**
Li-CNT	473–663 K (200–400°C)	0.1	0.72–4.2	**Pinkerton 2000**

Table 1. Summary of reported results achieved for hydrogen storage in carbon nanostructures. SWNT = single-walled carbon nanotubes, MWNT and CNT = multi-walled carbon nanotubes. Table references are appended following the endnotes to this report. (From Ding et al., Ref. 34)

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Early work that found up to 20 wt% hydrogen uptake by lithium-doped carbon nanotubes and up to 14 wt% by potassium-doped carbon nanotubes [40] was not reproducible, and it was later shown that most of the weight gain was due to water rather than hydrogen [41].

A “Review of theoretical calculations of hydrogen storage in carbon-based materials” was carried out by Meregalli and Parrinello in 2000 [42]. They drew two major conclusions: (1) “The reported calculations indicated a hydrogen uptake smaller than the more optimistic experimental results.” (2) “Furthermore the calculations suggest that a variety of complex chemical processes could accompany hydrogen storage and release.” In particular, they find both physisorption and chemisorption contributing to hydrogen uptake. They found potential for up to 12.5-wt% storage in GNFs, with most results below the 6-wt% target. For SWNTs, calculations gave a theoretically possible maximum of 14-wt% and for MWNTs, 7.7 wt%, relatively encouraging if such numbers can be achieved experimentally. However, the amount available for release could be small compared the amount stored.

Section Summary

The only proven system for hydrogen storage today that is practical for fuel cell vehicles is compressed gas high pressure tank storage. While this technology has the disadvantages of limited energy density and possibly high weight for the tank, it has been shown to workable with up to 70 GPa (10,000 psi) on-board storage tanks.

Of the other hydrogen storage systems, advanced carbon materials have been especially intriguing possibilities. However, very mixed experimental results have left us to conclude that these materials are far from proven to have adequate storage capacity. Further research is ongoing, but a breakthrough is needed to provide a foundation for confidence that carbon nanotubes or related materials will be able to satisfy storage requirements.

Finally, new ideas in hydridation of metal-N-H systems are sufficiently interesting that they are being pursued and may lead to the development of practical storage systems.

HYDROGEN FUEL STORAGE SAFETY

Hydrogen has a reputation for being explosive and therefore raises concerns about the safety of carrying a substantial quantity of H_2 in a vehicle fuel tank. However, because H_2 is the lightest gas, it has a tendency to diffuse away quickly in case its container is breached and consequently may represent less of a hazard than gasoline.

The simplest way to carry hydrogen fuel in a car or other vehicle is as a high-pressure gas 3-10 kpsi (21-69 MPa) in metal or composite-reinforced (fiberglass, carbon fiber, Kevlar) tanks. This is similar to the way compressed natural gas (CNG) vehicles operate.

There is an interesting report on H_2 for energy use [43] by the Norwegian environmental organization Bellona with useful safety information in Chapter 5. These authors conclude that "hydrogen is no more or less dangerous than any other energy carrier and furthermore that hydrogen has properties that in certain areas make it safer than other energy carriers: it is not poisonous, and has the ability to dissipate quickly into the atmosphere because of its light weight compared to air." They describe tests by Lockheed and Arthur D. Little that indicated that H_2 is, if anything, safer than gasoline or jet fuel. There are a number of references to crash testing by BMW [43, 44] that say BMW demonstrated the safety of H_2 fuel for cars.

Regarding the Hindenburg accident, a recent study [45] concluded that the paint on the dirigible skin was extremely flammable and was the true cause of the disaster. Of the 61 crew plus 36 passengers (total 97), 35 died, plus one person on the ground. Of the 35, 27 jumped when the Hindenburg was in the air and 8 were killed by burning diesel fuel [43,45]. There was no explosion. "The fire of the hydrogen from the gas cells lasted only less than one minute, and there is no evidence that anybody was directly hurt by it" [45].

Dr. Michael Swain at the University of Miami set fire to two cars, one carrying gasoline and the other hydrogen [46, 47]. The gasoline car had a 1/16" puncture in a fuel line. The hydrogen car had a leaking hydrogen connector. The gasoline-filled car was completely destroyed in an intense fire and the hydrogen car was essentially undamaged.

CONCLUSIONS

The efficiency of fuel cells in practical driving conditions can be 50-60%, but the overall efficiency (wells-to-wheels) is no more than 30% when production of H_2 from hydrocarbon resources is taken into account, significantly less than the FreedomCAR goal (60%×70% = 42%).[4] In comparison, the overall efficiency for the conventional internal combustion (IC) engine is about 15%. Hybrid-electric gasoline ICs are expected to reach almost 25%. On-site H_2 production by electrolysis from renewable energy sources is about 50% efficient, giving an overall fuel cell vehicle efficiency of 25%. Reforming natural gas is the most practical source of H_2 at present. Nuclear power production of hydrogen, however, may prove practical in the long term.

For fuel cell vehicles to be economically viable, the cost of fuel cell stacks must be reduced to ~$100/kW compared to the $1500/kW achieved thus far. Advances in catalyst (Pt) technology have significantly reduced expensive Pt usage. The FreedomCAR cost target is $45/kW.

Hydrogen storage is still a major research problem. While progress has been made, current systems are inadequate or marginal. On-board storage of sufficient H_2 most likely will be as compressed gas. Demonstration vehicles have used 5000-psi carbon-fiber wrapped tanks, and tanks have been certified recently at pressures of 10,000 psi. Liquid hydrogen and cryoadsorption storage are almost certainly not practical for vehicle use. Metal hydridation and metal-N-H systems show some promise, but much research is still required for these systems. Advanced carbon materials are intriguing, but contradictory research results, with relatively low reproducible storage capacities, have been discouraging and much remains to be done. Unresolved research questions, such as hydrogen storage, make the fuel cell vehicle approach a long-term issue. It is not a short-term solution to energy or emissions problems.

For the next twenty years, vehicles powered by the hybrid-electric internal combustion engine could well prove to be just as beneficial, from a life-cycle perspective, as fuel cell vehicles. A recent MIT study [21] finds that fuel cell vehicles do not have significant advantages (in terms of energy consumption or CO_2 emissions) over future vehicles powered by hybrid-electric, internal combustion engines.

Obviously no widespread infrastructure exists, but distribution might not be as daunting as first thought. Several pipelines for H_2 transmission already exist and natural gas lines could be retrofitted for these purposes. Transportation by tanker truck over short distances is well established. The FreedomFUEL initiative recently announced by President Bush is intended to address some of the infrastructure and cost issues. Realistic estimates of when substantial market penetration of fuel cell vehicles will occur (at least 15 years) allow plenty of time to develop an infrastructure. Scenarios of how a hydrogen society might evolve have been explored by Burns *et al.* [14] and Chinworth [48].

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[4] Hopefully, the sources of energy for H_2 production will eventually be renewable.

The motivation for the FreedomCAR and FUEL initiatives is to reduce US petroleum imports, greenhouse gas emissions, and atmospheric pollutants. This study shows that hydrogen fuel cell vehicles as well as advanced internal combustion hybrid electric vehicles can accomplish significant reductions in energy usage by the transportation sector with the concomitant reduction of CO_2 emissions. Roughly, a factor of two reduction in life-cycle energy (and CO_2) per unit of distance traveled relative to today's vehicles is possible. The emissions of atmospheric pollutants such as CO and NO_x depend strongly on the source of hydrogen, however. Only generation of hydrogen by renewable energy sources or by nuclear power truly reduces emissions of pollutants, although the point of origin may be away from urban areas. The FreedomCAR initiative includes more than just fuel cells, which is fortunate because it is not clear what technology may be the best for the future.

Nonetheless, there was optimism at a recent DOE workshop [53] that, despite daunting challenges, many good ideas remain to be pursued. Research carried out for hydrogen initiatives could well improve long-term energy security.

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General Disclaimer
This POPA E&E Subcommittee report represents only the views and estimates of individual members and contributors and not necessarily of their organizations.

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The California Air Resources Board at www.arb.ca.gov/homepage.htm

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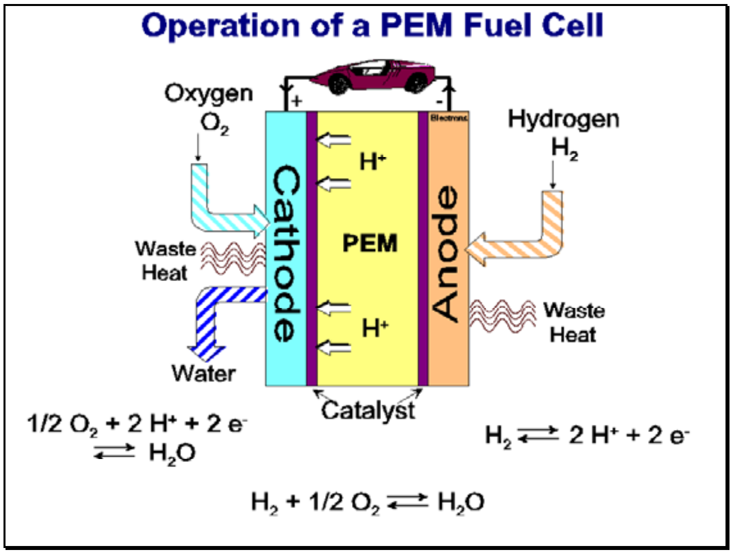
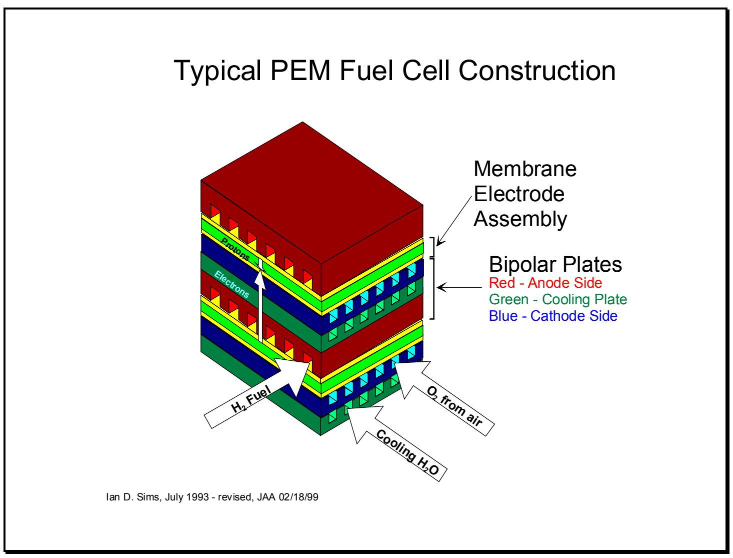
APPENDIX A
From Ref. [8].



Principles of operation.



Schematic of a fuel cell.

APPENDIX B

FreedomCAR vision and goals [2]:

Vision

Affordable full-function cars and trucks that are free of foreign oil and harmful emissions, without sacrificing safety, freedom of mobility, and freedom of vehicle choice.

Technology-Specific 2010 Goals[1]

- To ensure reliable systems for future fuel cell powertrains with costs comparable with conventional internal combustion engine/automatic transmission systems, the goals are
    - Electric propulsion system with a 15-year life capable of delivering at least 55 kW for 18 seconds and 30 kW continuous at a system cost of $12/kW peak.
    - 60% peak energy-efficient, durable fuel cell power system (including hydrogen storage) that achieves a 325 W/kg power density and 220 W/L operating on hydrogen. Cost targets are $45/kW by 2010, $30/kW by 2015. [2]

- To enable clean, energy-efficient vehicles operating on clean, hydrocarbon-based fuels powered by either internal combustion powertrains or fuel cells, the goals are
    - Internal combustion systems that cost $30/kW, have a peak brake engine efficiency of 45%, and meet or exceed emissions standards. [The term brake engine efficiency refers to output shaft measurements.]
    - Fuel cell systems, including a fuel reformer, that have a peak brake engine efficiency of 45% and meet or exceed emissions standards with a cost target of $45/kW by 2010 and $30/kW in 2015. [2, 3]
- To enable reliable hybrid electric vehicles that are durable and affordable, the goal is
    - Electric drivetrain energy storage with 15-year life at 300 Wh with discharge power of 25 kW for 18 seconds at a cost of $20/kW
- To enable the transition to a hydrogen economy, ensure widespread availability of hydrogen fuels, and retain the functional characteristics of current vehicles, the goals are
    - Demonstrated hydrogen refueling with developed commercial codes and standards and diverse renewable and non-renewable energy sources. Targets: 70% energy efficiency well-to-pump; cost of energy from hydrogen equivalent to gasoline at market price, assumed to be $1.25 per gallon (2001 dollars). [4]
    - Hydrogen storage systems demonstrating an available capacity of 6 wt% hydrogen, specific energy of 2000 W-h/kg, and energy density of 1100 W-h/L at a cost of $5/kWh. [5]
    - Internal combustion systems operating on hydrogen that meet cost targets of

$45/kW by 2010 and $30/kW in 2015, have a peak brake engine efficiency of 45%, and meet or exceed emissions standards.

- To improve the manufacturing base, the goal is
  - Material and manufacturing technologies for high-volume production vehicles that enable and support the simultaneous attainment of
    - 50% reduction in the weight of vehicle structure and subsystems,
    - affordability, and
    - increased use of recyclable/renewable materials.

Notes:

1. Cost references are based on CY 2001 dollar values. Where power (kW) targets are specified, those targets are to ensure that technology challenges that would occur in a range of light-duty vehicle types would have to be addressed.

2. Does not include vehicle traction electronics.

3. Includes fuel cell stack subsystem, fuel processor subsystem, and auxiliaries; does not include fuel tank.

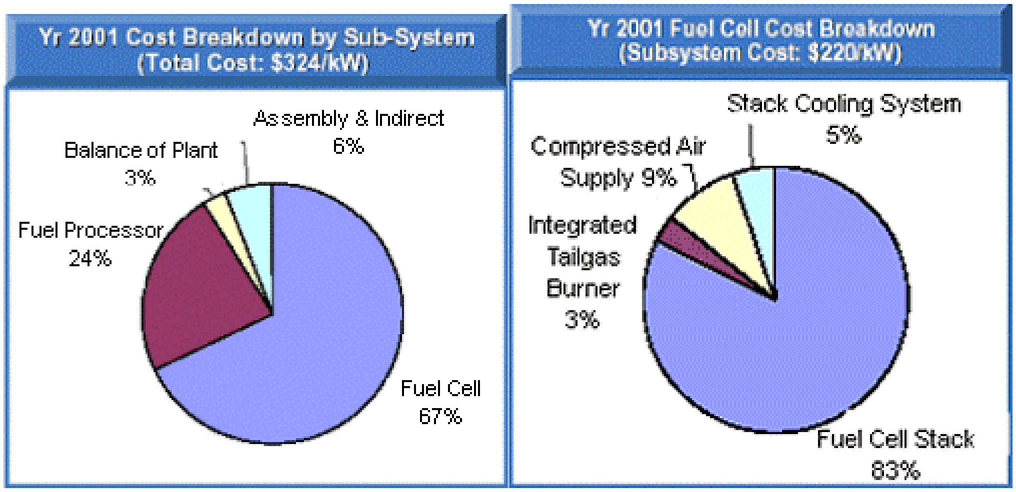
4. Targets are for hydrogen dispensed to a vehicle assuming a reforming, compressing, and dispensing system capable of dispensing 150 kg/day (assuming 60,000 SCF/day of natural gas is fed for reforming at the retail dispensing station) and servicing a fleet of 300 vehicles per day (assuming 0.5 kg used in each vehicle per day). Targets are also based on several thousand stations, and possibly demonstrated on several hundred stations. Technologies may also include chemical hydrides such as sodium borohydride.

5. Based on lower heating value of hydrogen; allows over a 300-mile range.

APPENDIX C

From Ref. [49].

Reformer/Fuel Cell Cost Estimates				
	Annual Production Rate			
	500	**10,000**	**30,000**	**500,000**
Fuel Cell Stack	$17,258	$9,618	$9,394	$8,509
Air Loop	$1,160	$821	$734	$529
Water Loop	$1,106	$832	$757	$605
Coolant Loop	$620	$486	$450	$386
ATR	$3,531	$1,945	$1,532	$1,322
Reformate Loop	$1,172	$838	$739	$658
Fuel Loop	$879	$616	$573	$466
Controls	$719	$501	$442	$316
Misc./BOP	$320	$240	$220	$150
System Assembly	$723	$487	$442	$157
Total Cost	$27,489	$16,384	$15,282	$13,099
Cost/kW	$550	$328	$306	$262
Based on a 50kW-net system. All costs are preliminary, as DFMA optimization had not yet been completed. ATR and fuel cell stack examined in more detail than other system components. All costs include 10% cost contingency and markup to reflect profit, G&A.				



From Ref. [50].

<u>Argonne National Lab Estimate for Central Production Facilities</u> (quoted by Prof. Daniel Sperling, U. of California, Davis [51]):

- Infrastructure cost for producing H_2 equivalent to 1.6 x 10^6 barrels/day of oil (~20% of current automobile and light truck usage.)
- $400 billion for production
- $175 billion for distribution

[For comparison, the Exxon-Mobil market capitalization is $200 billion.]

<u>BP Company Estimate for Outfitting Local Stations with Natural Gas Reformers</u>

- $400,000 for reformer to make hydrogen using local gas lines
- $1.5 million to build conventional gas station

From Ref. [52].

APPENDIX D

Energy consumed in MJ/km (1 MJ/km = 1500 BTU/mi is equivalent to 0.013 gal/mi of gasoline.)

	2001 gasoline ICE	2020 gasoline ICE HEV	2020 diesel HEV	Fuel cell vehicle
Vehicle operation	2.47	1.07	0.92	0.54
Fuel cycle	0.52	0.22	0.13	0.42
Manufacturing	0.29	0.26	0.26	0.28
Total	3.28	1.55	1.31	1.24

[From Tables 8 and 9 of Ref. 21.]

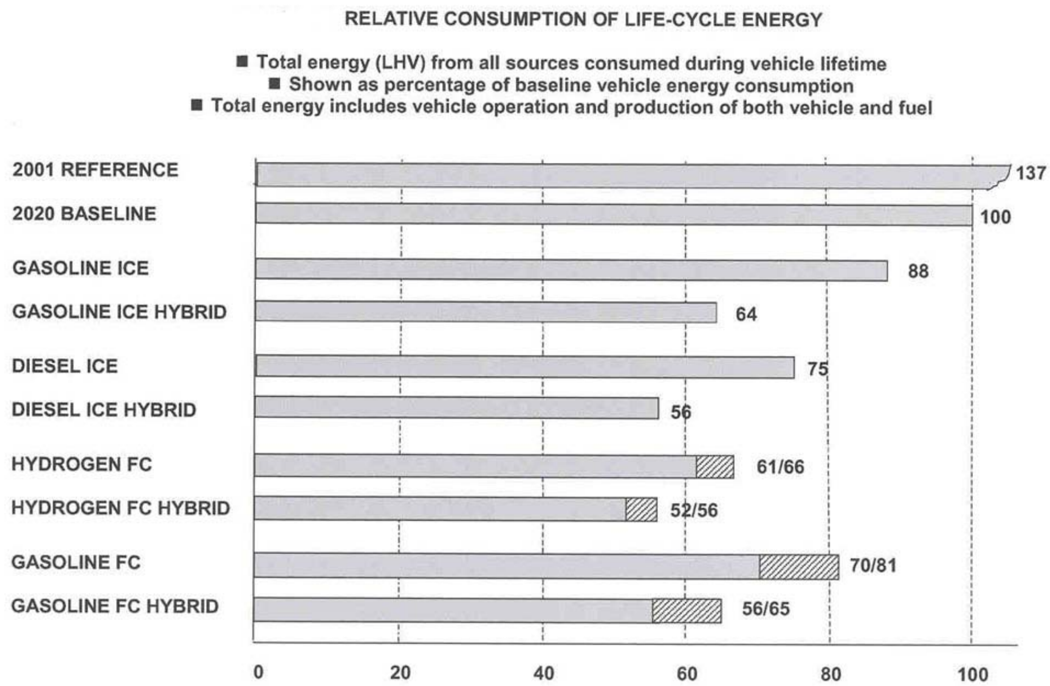
The predicted well-to-wheels energy consumption for the 2020 fuel cell vehicle is 0.96 MJ/km = 1400 BTU/mi which is considerably less than the estimate in Table I. This reflects different assumptions about future technology development. For the conventional gasoline engine, the results also differ—4500 BTU/mi (3.0 MJ/km) compared to 6500 BTU/mi. The later calculation assumes a 22-mpg vehicle compared to 31 mpg here.

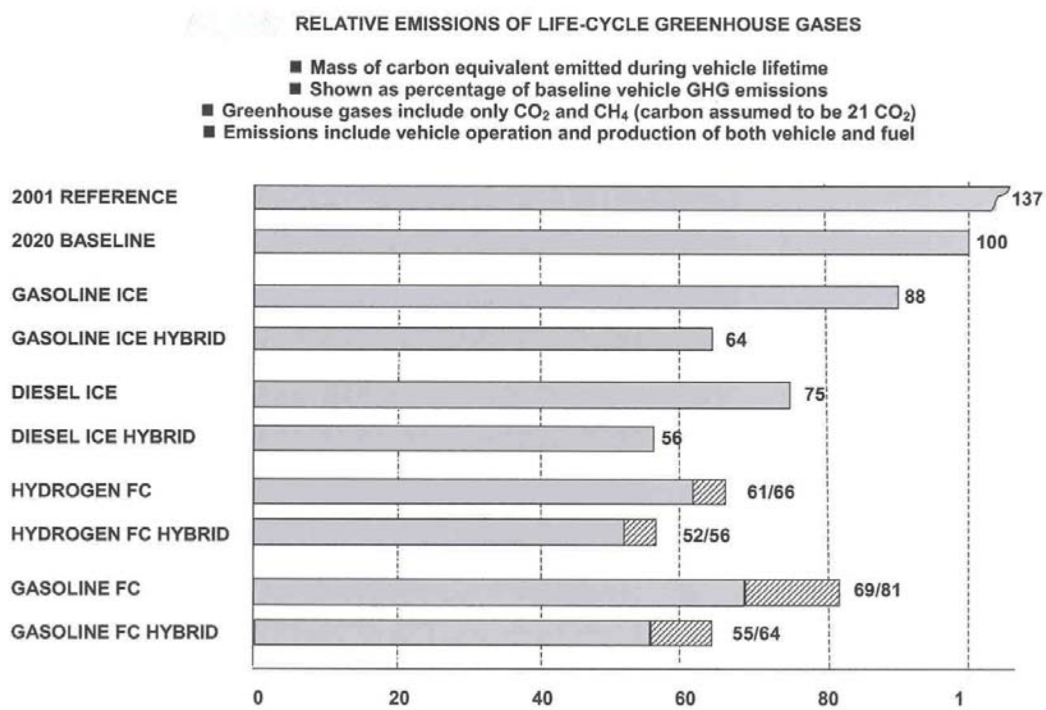
Life-Cycle Greenhouse Gas Emissions (gC/km).

	2001 gasoline ICE	2020 gasoline ICE HEV	2020 diesel HEV	Fuel cell vehicle
Vehicle operation	48.5	21.0	19.1	0
Fuel cycle	12.1	5.2	3.0	19.4
Manufacturing	5.5	5.0	5.1	5.3
Total	66.1	31.2	27.2	24.7

[From Tables 8 and 9 of Ref. 21.]

The emissions of greenhouse gas (GHG) in grams of carbon per kilometer traveled as given in Ref. 21 shows the significant improvements expected from advanced ICE HEVs and fuel cell vehicles. Life-cycle GHG emissions are approximately 20 g/MJ for the four vehicles tabulated in the two tables above.


RELATIVE CONSUMPTION OF LIFE-CYCLE ENERGY
■ Total energy (LHV) from all sources consumed during vehicle lifetime
■ Shown as percentage of baseline vehicle energy consumption
■ Total energy includes vehicle operation and production of both vehicle and fuel
2001 REFERENCE
137
2020 BASELINE
100
GASOLINE ICE
88
GASOLINE ICE HYBRID
64
DIESEL ICE
75
DIESEL ICE HYBRID
56
HYDROGEN FC
61/66
HYDROGEN FC HYBRID
52/56
GASOLINE FC
70/81
GASOLINE FC HYBRID
56/65
0
20
40
60
80
100


RELATIVE EMISSIONS OF LIFE-CYCLE GREENHOUSE GASES
■ Mass of carbon equivalent emitted during vehicle lifetime
■ Shown as percentage of baseline vehicle GHG emissions
■ Greenhouse gases include only CO_2 and CH_4 (carbon assumed to be 21 CO_2)
■ Emissions include vehicle operation and production of both vehicle and fuel
2001 REFERENCE
137
2020 BASELINE
100
GASOLINE ICE
88
GASOLINE ICE HYBRID
64
DIESEL ICE
75
DIESEL ICE HYBRID
56
HYDROGEN FC
61/66
HYDROGEN FC HYBRID
52/56
GASOLINE FC
69/81
GASOLINE FC HYBRID
55/64
0
20
40
60
80
1

From Ref. [21].