

Fuel cell technology and innovation

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Abstract

Fuel cells are an innovative technology, but present designs require much more innovation before they can make a large impact on future energy conversion markets. They are regarded today as competition for dispersed generation using conventional clean fossil fuels. Since they use hydrogen, they are built into large chemical plants which may be regarded as dispersed refineries. The size of the plant is determined by this refinery, which cannot be scaled down effectively to the units of a few kW or less, where the fuel cell potentially excels. If they are liberated from this requirement either by the use of new materials or (more likely) by the direct use of hydrogen, first from natural gas, then from renewables, they can have a major impact. How innovation may be used to tackle the home and automobile energy conversion markets is considered.

1. Introduction

Compared with thermal engine approaches to chemical energy conversion, fuel cell technology is most certainly innovative. Thermal engines use the heat generated by unconstrained chemical reaction to expand a working fluid, which can then be cooled and contracted, thus performing mechanical work. This operation is subject to the well-known Carnot conversion limitation [1]. A fuel cell performs direct conversion of potential energy (chemical free energy) available in a chemical reaction to direct current electrical work, which can then be transformed into mechanical work at almost 100% efficiency by means of an electromagnetic device, the electric motor. In both cases, the chemical reaction is normally, but not invariably, the oxidation (by oxygen) of a suitable fuel. For terrestrial use, the oxygen is normally from the atmosphere.

Any device which converts one form of energy into another more useful form in a single step is loosely said to operate by direct energy conversion. Examples are thermionic or thermoelectric converters of heat to electrical work, or photovoltaic cells which transform photon beams into electrical work. Both of the former are Carnot-limited heat engines. The latter is only a true direct converter of one form of work to another if the photon beam is in a very narrow energy range, as in a laser. If the photons are from an incandescent object as a heat source, they become the thermodynamic working fluid between it and the photovoltaic cell acting as the heat sink. The whole system is then a Carnot-limited heat engine. Thus, solar photovoltaic and solar thermal devices are thermodynamically equivalent.

Gravitational potential energy (e.g., falling water) is mechanical work, and it may be converted into electricity using the electric motor operating in reverse as a generator. In such a device, the falling (conductive) liquid could be the working fluid in a magnetohydrodynamic (MHD) machine. It is instructive to note that all three concepts

were discovered by Michael Faraday. In the motor and generator, whose principles he discovered in 1821 and 1831, respectively, a moving, i.e., rotating, electronic conductor moves through a magnetic field, which is typically fixed. In MHD, the magnetic field is again fixed, but the conductor is ionic, and requires electrodes in contact with it at which electrochemical reactions take place to complete the circuit. Unlike the rotating electronic conductor in the typical motor or generator, the MHD conductor is normally a linear free-flowing stream. Faraday's first MDH experiment to detect an electromotive force in a moving conducting fluid used the earth's fixed magnetic field, with two electrodes immersed in the Thames from Waterloo bridge in December 1832. In proposed MHD devices for power generation, a rapidly-moving ion-seeded conducting plasma produced by a rocket motor or an explosive device is employed as the ionic conductor, rather than a slow-moving poorly-conducting river. Since the plasma flow clearly is a thermal working fluid, the whole system is again Carnot-limited.

2. Fuel cell energy conversion

Compared with other methods of energy conversion, the fuel cell (indeed any electrochemical cell or battery) is a unique and innovative device which converts a relatively inaccessible form of potential energy, i.e., chemical free energy of reaction, directly into work without any express or implied heat engine limitations.

The electrodes in an electrochemical cell release electrons whose energy corresponds to the chemical potential (free energy) change in the fuel-oxidant reaction. A simultaneous ion stream, which is equal and opposite to that of the electrons, must also occur in a separate pathway. In principle, either stream can do work using a concentration gradient. The ion gradient can perform a chemical reaction, or can allow the transport of solvent such as water, thereby acting as a pump. Since we can carry out these objectives better by electrolysis or electrical work, a practical electrochemical cell is designed to eliminate ion gradients, and operates on the electron gradient, i.e., on the electromotive force. Nature operates via microscopic fuel cells which convert oxidizable species (sugars and their derivatives) and oxidizing species (oxygen and oxidizing redox derivatives) into work via ion (proton) gradients across membranes between her reaction sites or electrodes in the mitochondria of living cells [2]. Nature has largely ignored the possibility of using gradients of electrons, transmitting the latter between her electrodes via redox systems which act as a shorted copper wire.

Whereas nature uses partially oxidized organic fuels, e.g., glucose, hydrogen is the most active common fuel for technical fuel cells today. Under the same experimental conditions, it has an oxidation rate perhaps four orders of magnitude greater than that of simple partially oxidized one-carbon compounds such as methanol, which in turn oxidize faster than saturated hydrocarbons by about the same amount. Among nitrogen compounds carrying hydrogen, ammonia is about as active as methanol, whereas hydrazine is an active, but impractical, fuel. In consequence, today's electric utility or on-site co-generation fuel cells operate on hydrogen derived from common fuels such as natural gas, which must receive chemical processing before use. The size and cost of this chemical plant, which must be thermally integrated with the electrochemical cell for maximum efficiency, severely limits fuel cell application, since it is likely to represent 60% or more of total system cost, weight and volume. If hydrogen fuel becomes available from future renewable, nuclear, or other sources, the fuel cell

generator will become a much more attractive proposition, since it will have a lower capital cost.

Nature's living machines operate on carbohydrates at a maximum of few W/kg (or W/l), and then only for short periods, and at a maximum net thermal efficiency of about 30% after accounting for parasitic energy requirements to operate metabolism and circulation. Certain fuel cells today can operate on pure hydrogen and oxygen at a specific power of better than 2000 W/kg without auxiliary systems (controls and cooling), or 1300 kW/kg net, at a thermal efficiency of 55%. These specific power outputs are projected to more than double in future. In comparison, internal combustion (IC) racing car engines operate at 4000 W/kg at slightly over 20% efficiency, whereas the family car IC power plant may offer 500 to 750 W/kg, depending on its technology. A steam locomotive of earlier this century would certainly have produced less than 10 W/kg at less than 10% efficiency, whereas more efficient and complex condensing steamship power plants (about 20% efficiency) had even lower power densities. Large steam plants today (35% or more efficiency) have specific power outputs which are similar, i.e., they require a large amount of construction material to furnish their power output.

In comparison, military jet engines may operate at close to 10 000 W/kg (10 kW/kg) at 30% efficiency. However, these engines are air-breathing, whereas the above fuel cell uses pure pressurized oxygen, like a rocket motor. If the same hydrogen fuel cell were air-breathing, its power density in kW per unit active area, or kW/kg, would fall by a factor of five or more at a similar overall efficiency. An attainable figure today would be about 250 W/kg for the whole system, which might be projected to provide 1 kW/kg in future as lighter designs are adopted.

The fuel cell has not generally been considered to be a practical power source for aircraft, because its product electricity must be converted into motion using a relatively heavy electric motor, for example to power a propeller. The rocket motor is similarly at a disadvantage if its energy product must be electricity rather than motion. In principle, a hydrogen-oxygen rocket with a steam turbine, condenser and electrical generator (or alternatively an MHD generator), can also operate as 55% efficiency heat engine. However, it cannot do this with the specific power rating of a fuel cell which directly produces d.c. electric power. For this application, the hydrogen-oxygen fuel cell is therefore a better choice than a closed Rankine-cycle hydrogen-oxygen thermal engine.

3. Fuel cell system end-use optimization

That any energy conversion system must be optimized from the fuel-oxidant combination to the desired work output applications seems obvious, but it is often forgotten. To avoid the use of an integrated chemical plant, a fuel cell generator must operate on hydrogen fuel. It is then an efficient, high power density device, thanks to the effectiveness of today's hydrogen anodes. These show very high hydrogen oxidation rates, so they are essentially thermodynamically reversible even at net current densities of 1 A/cm² or greater for the best systems operating close to ambient temperature. For certain applications which use hydrogen fuel and where light weight is desired, the direct use of the d.c. power output of a fuel cell may be convenient. This avoids the use of d.c. to a.c. conversion equipment, so that the whole power plant becomes the fuel cell stack and its associated auxiliaries, which include the cooling and product

water removal systems. In general, these will have a weight and volume which is less than that of the stack itself, so that the generator is small and compact so long as the hydrogen fuel is freely available from an external supply, e.g. a pipeline.

If the generator must be either transportable or mobile, then the weight and volume of hydrogen storage must be considered, depending on the mission anticipated. If the power device must perform traction work, particularly in demanding applications such as an air flow for an aircraft, the extra weight and volume of an electric motor and, for example, a propeller must be added. This and the requirement for hydrogen storage makes a fuel cell prime mover much less desirable, unless the power requirement is modest and the mission is very long, so that fuel weight and volume savings become important. If the mission for an aircraft is so long that fuel requirements are large compared with the motor weight and volume, the use of (liquid) hydrogen fuel may be considered. For a given energy content, it would have only one third of the weight of a hydrocarbon fuel, but unfortunately about three times the volume, due to its low density. It also requires a suitable complex cryogenic tank, whose relative weight will be prohibitive if the quantities of liquid hydrogen are small, but whose weight relatively diminishes with increasing quantities of hydrogen and thus surface-to-volume ratio. We should note that a good metallic hydride or compressed hydrogen or metallic hydride storage tank will only store about 1.5 wt.% of hydrogen, and would have two to six times the volume of liquid hydrogen. These figures would be independent of the quantity of hydrogen stored. The storage subsystem may then be about twenty times heavier per unit of energy than a hydrocarbon, and would occupy about six to twenty times the volume. Certainly for aerospace use, only liquid hydrogen, or the slightly denser 'slush' (solid-in liquid) hydrogen can therefore be considered.

Thus, for a high-payload rocket requiring a large amount of fuel (for example, the main tank and engines of the Space Shuttle), liquid hydrogen is attractive, since savings in fuel weight increase payload. Fuel volume is not very important, since high speed is achieved outside the dense atmosphere. For use in the atmosphere, the aerodynamic drag of increased fuel volume with a shape of low aspect-ratio must be traded off against the lower weight, therefore the possibility of using smaller and lighter aerodynamic surfaces. If there are other reasons for choosing liquid hydrogen fuel, for example its heat sink capacity in a hypersonic vehicle, its use will be doubly advantageous, provided that the vehicle is not volume-limited, i.e., for an aircraft shape corresponding to a low-aspect-ratio lifting body. This is the case for the National Aerospace Plane (NASP).

An example of the proposed use of a liquid hydrogen fuel cell prime mover in a relatively conventional low-speed long-wing aircraft is for a high-altitude surveillance drone with circumnavigational range. Since the fuel cell may be twice as efficient as an IC engine operating under optimal conditions, the corresponding liquid hydrogen fuel weight would then be one-sixth of that of gasoline, with a volume only about 50% higher. The aircraft can take off like a glider, and requires a low cruise power, hence the fuel cell and electric motor weight is small. The fuel cell must be air-breathing, so for ultra-high-altitude flight (30 km) another problem arises, namely the requirement for compression work to produce an effective fuel cell power density in rarified air. The waste energy from most lightweight fuel cells is in the form of heat at almost ambient temperature, so this must be provided by parasitic electricity. This increases weight, and degrades net efficiency from 55% to 40% in a small fuel cell system, whereas the overall efficiency of a competing IC engine for which air compression is provided by high-temperature exhaust heat may increase somewhat under these operating conditions, for example from less than 25% to approaching 30%.

Thus, the advantages of fuel cell electric drive for low speed, long-range flight as high altitude start to become marginal, especially if the IC engine is liquid-hydrogen-fueled. Its use can only be justified if the power requirement is perhaps 5 kW or less, when the efficiency of the thermal engine starts to diminish. Finally, the most efficient and highest power density hydrogen fuel cells, namely alkaline systems, today require a chemical scrubber to remove carbon dioxide from ambient air, which further increases weight and somewhat reduces efficiency.

At first sight, the fuel cell seems to have major advantages over its rival thermal engines for use under atmospheric pressure conditions. It is very efficient, and has the potential to become more so as knowledge of electrocatalysis is extended. Like any other battery, it consists of identical units of equal efficiency, which can be scaled up or down as required with unchanged performance. A submicron fuel cell with 50% efficiency could be made, if required. Again like any common battery, it is silent and pollution-free in use, and is a passive block which should require no internal maintenance. Unlike many batteries, which may use chemical reactions involving compounds posing some hazard if released to the environment, its only product is pure water. Philosophically, it seems to be the perfect way of conversion of chemical energy to work in its most universal form, electricity. It represents the direct connection between chemistry and physics. Thus, why is it not being proposed to replace the IC engine?

4. Fuel cells versus IC engines for passenger vehicles

An ambient-air fuel cell operating on hydrogen, even if inexpensive and well designed, will barely compete with the family car engine for terrestrial transportation in terms of power density. More particularly, it must compete in terms of capital cost, effective lifetime, cost, availability, and practicality of fuel, and finally on capital cost.

The mass-produced IC engine should have a life of about 5000 intermittent operating hours if correctly maintained. A suitable fuel cell should compete with the automobile IC engine in regard to lifetime. For the Space Shuttle Orbiter alkaline unit [3], whose man-rated design was essentially frozen in 1973 and is therefore the oldest fuel cell system still in regular use, no maintenance is required for 2500 h, when the stacks are replaced as a safety measure for the high-risk orbital application. At the same time, certain components are overhauled. At 5000 and 7500 h, stacks are replaced, and progressively more extensive overhaul occurs. Space Shuttle Orbiter cell components have operated on the laboratory bench for 100 000 h. The International Fuel Cells (United Technologies) phosphoric acid fuel cell stacks are designed to operate for 40 000 h without maintenance, and again, their older-generation components have operated on the laboratory bench for over 100 000 h [4]. The above systems have liquid electrolyte, whose retention inside the cell must be carefully controlled, and which can be a critical item in determining cell lifetime as other changes take place, for example slow oxidation of cathode components, which leads to increased wettability and electrolyte redistribution. The acid fuel cell with a fluorinated solid polymeric sulfonic acid electrolyte (the proton exchange membrane or PEM system) has a stable electrolyte which is itself immobilized, and it has shown very long lifetimes (more than 40 000 h using early components) with very low degradation [5]. As in the case of IC engines, components which limit fuel cell system lifetime are likely to be mechanical.

Unleaded gasoline at the refinery is 67 ¢/gallon (US) today, and 90 ¢/gallon, or about \$6.80/GJ, as untaxed fuel at the pump. Hydrogen made from natural gas at

today's low prices (less than \$1.50/GJ) in the United States would cost about \$6.00/GJ at a large central plant. This would have a capacity of 2.8×10^6 m³/day of hydrogen (100 million SCF/day), or about 185 MW equivalent, assuming 50% conversion efficiency of the hydrogen in fuel cells (4.4 GW h/day). On a five day per week basis, this would be enough to supply about one million 1 tonne commuter cars, each using 0.11 kW h/tonne-km at the motor terminals, for a 55 km roundtrip each weekday. The circulation assumes some storage capacity associated with a pipeline distribution system in a relatively large urban and suburban area of perhaps 1000 km².

Transporting gaseous hydrogen is as technically feasible as transporting natural gas, although it would be somewhat more expensive because of the greater volume required for a given amount of energy. However, the hydrogen pipeline infrastructure does not presently exist. It did exist in many parts of Europe for the transport of coal gas, but this has largely been converted and extended or replaced by a natural gas distribution system. Thus, the manufacture and distribution of hydrogen for a population of 1 million commuter vehicles would require a considerable investment, in the order of \$100–200 million. The annual capital charges on this sum, representing the central plant plus the distribution system, will only represent less than \$20 per vehicle per year. It is therefore feasible, and the technology certainly requires no breakthroughs. However, there are institutional impediments connected with building up the infrastructure. One impediment is whether it is even desirable to replace a congestive, polluting technology for commuter vehicles by one which is non-polluting, but equally congestive. The future may indeed require less commuting, if 'telecommuting' electronic technologies enable people to work at home.

Assuming that a system is put in place, the cost of hydrogen at the distribution station, based on \$10 per vehicle-year for distribution costs, or less than \$1.00/GJ, might be \$8.00/GJ including profits, but before taxes. However, hydrogen consumption will be about three times less than for an IC engine for a given range in urban use if a 50% efficient fuel cell is used. This estimate is based on the higher heating value of the hydrogen, i.e., for a fuel cell operating at an average of 0.74 V, and the measured energy requirements of electric vehicles. The cost per km for the consumer would therefore be 40% of that for gasoline. The entire system would have essentially zero emissions. It could therefore represent a great social opportunity, and prepare for a non-fossil hydrogen future.

The final questions are those associated with hydrogen storage, and fuel cell cost. The first is discussed in the next two sections. Discussions of the costs of stationary utility and vehicle fuel cells follow after consideration of fossil fuels as feedstock.

5. Hydrogen storage

As in the case of aircraft, the problem of hydrogen use in a vehicle is the method of storage. Cast-iron cylinders store considerably less than 1 wt.% of compressed hydrogen. Cylinders may eventually be able to store 3 wt.% hydrogen if they are made from suitable high-strength materials, for example with external composite reinforcement. Such pressure vessels may be costly, and they will allow less than 10% of the range per total weight of fuel plus container if an IC engine is intended to operate on compressed hydrogen instead of gasoline.

However, to attain the same urban ranges for hydrogen (in the fuel cell vehicle) and gasoline (with an IC engine vehicle with similar specifications) would require a

lightweight pressure cylinder only about three times heavier than that of the corresponding weight of gasoline. The weight using more common storage cylinder materials will be appreciably greater than this. We should note that for a given cylinder material, weight is essentially independent of pressure, whereas volume is inversely proportional to it. For storage cylinders operating at 200 bar pressure (about 3000 psi), the storage volume required to give a hydrogen fuel cell vehicle the same range as that of a gasoline vehicle will be about five times greater than the volume of gasoline required. However, gasoline tanks can occupy space as desired, whereas conventional pressure vessels must be spherical or cylindrical with spherical end caps. This results in wasted space equal to 60% or so of the volume of stored compressed gas, increasing the required volume to eight times that of gasoline.

In support of the use of compressed hydrogen, we should point out that compressed natural gas is often advocated for use in IC engines, and natural gas or other clean fuel was mandated by law in Texas for public vehicle fleets and larger school bus fleets starting in 1994 [6]. On July 31, 1990, propane and electricity were included in the definition of clean fuel. Hydrogen has 30% of the energy per unit volume as natural gas. Hence compressed natural gas (using the IC engine) and hydrogen (using a fuel cell) should give about the same urban range per unit volume and weight. If the public and vehicle manufacturers can be persuaded that natural gas is acceptable, then hydrogen should also be acceptable using identical pressurized storage equipment.

Another system for storage of hydrogen under moderate pressure is via a reversible metal hydride. If materials such as $\text{Fe}_{0.9}\text{TiMn}_{0.1}$ [7] are used, a typical mass of hydride (including the moderate-pressure container) will store up to about 1.5 wt.%, making this option less attractive than a high pressure lightweight cylinder. Hence, compared with the weight of gasoline, about five times the weight of metallic hydride alone may be needed for a fuel cell vehicle to give the same vehicle range as the IC engine. The corresponding volume within the pressure vessel containing hydride may be divided by as much as a factor of three compared with that of compressed hydrogen at 200 bar. This volume will depend somewhat on the porosity of the metal hydride. However, the system also requires heat exchangers, which can be incorporated in the surrounding dead volume, but they do increase system weight. As a result, the total system volume required for a given range in a fuel cell vehicle may be about four times that of gasoline. The disadvantage of metallic hydride storage will be weight (about equal to compressed storage in a simple cylinder holding 1 wt.%), and cost. The hydride materials now cost about \$30/kg retail, with about \$12/kg projected for future bulk cost, i.e., about \$700 per kg of hydrogen stored. This is too expensive for widespread applications.

Other options for hydrogen storage are as an absorbed compressed gas on high-surface area carbon at low temperatures ($-120\text{ }^\circ\text{C}$), and as a cryogenic liquid. The former is receiving attention at present [8]. The low density of the absorbant makes it advantageous compared with metallic hydrides by about a factor of six from the standpoint of absorbant weight, since 9.5 wt.% of hydrogen storage is claimed. The net hydrogen storage capacity might be 4 wt.% of the total weight of the absorbant, the lightweight insulated 55 bar pressure-vessel (an aluminum liner with a carbon fiber-epoxy outer shell) and the heat-exchanger. Hence the weight of the tank and heat exchanger to achieve the same urban range might be 2.5 times that of gasoline in the IC engine vehicle. The volume may be about the same as, or somewhat greater than, that of a corresponding metallic hydride bed. The cost of the absorbant may be quite low, perhaps one-tenth of that of a metallic hydride. The system is presently difficult to evaluate, since the weight, cost and volume of the cryogenic system required

is at present unknown. The system was designed to operate at the lowest working temperature of CFC-12, so it may require future revision.

The use of cryogenic hydrogen opens up a different set of issues. Evidently, it would be attractive from the weight and volume viewpoint. Approximately the same volume of liquid hydrogen would give the same range in a vehicle with a fuel cell as that of gasoline in one with an IC engine. The weight of the cryogenic tank would exceed the weight of hydrogen stored in amounts sufficient for a small vehicle, but by a factor of about 5, rather than by a factor of 30 or more for compressed hydrogen storage. The volume would however still be considerable. For relatively small quantities of hydrogen, the multiply-insulated Dewar vessel would have a volume more than twice that of the stored cryogenic liquid. Since the vessel would be cylindrical, the total volume occupied would be about three times that of a gasoline tank to give the same range with an IC engine.

Daimler-Benz [9] has provided some details of storage schemes for 145 kW h (higher-heating-value) of hydrogen, i.e., about 3.7 kg. This quantity of hydrogen should allow a one tonne fuel cell vehicle with 50% efficiency and state-of-technology rotating parts and drag coefficient to operate for 650 km either in an urban environment (0.11 kW h/tonne-km), or at a steady 90 km/h (10 kW cruise). A vehicle such as the General Motors 'Impact' (0.070 kW h/tonne-km) would have a correspondingly improved range. As a compressed gas at 300 bar (50% higher than the value assumed earlier, giving a lower volume), the system would occupy 250 l overall, and weigh 120 kg assuming a lightweight pressure vessel storing 3 wt.%. As a metal hydride enclosed in a smaller 50 bar pressure vessel, the corresponding figures for the entire subsystem would be 170 l and 320 kg, and as 40 l of cryogenic liquid in a 4 bar Dewar container, they would be 140 l and only 20 kg. As absorbed hydrogen on high-surface carbon, we may estimate corresponding figures of 200 l (?) and 110 kg. The weight figure for cryogenic hydrogen is a dramatic reduction from those for the other technologies, but the average vehicle is more restricted in volume than in weight.

Using gasoline and assuming three times the corresponding energy requirement per km under urban conditions (i.e., 6.9 l/100 km or 33.8 mpg), 45 l (11.9 US gallons) of gasoline would be required to give approximately the same urban range. Including the tank (50 l overall), this gasoline would weigh about 40 kg. We should remark that the above values for electric vehicles are based on actual measurement, whereas those for the gasoline vehicle are optimistic.

6. Cryogenic hydrogen and innovations in storage

If cryogenic hydrogen is advantageous from the viewpoint of weight, it is unlikely to be from the standpoint of purchase price, the capital cost of the plant required to produce it, or the energy requirement for liquifaction. The latter will add about 50% to the total primary energy requirement if conventional liquifaction is used, increasing the primary energy used per mile by this amount. This energy, in the form of electric work, may of course be supplied using primary electrical power from nuclear or future solar energy, not from the natural gas which will initially be used to manufacture hydrogen gas at about 70% thermal efficiency. Again based on present technology, the capital cost of the liquifaction plant will be higher than that of the plant to manufacture gaseous hydrogen from natural gas, giving a product which may cost at least a factor of two more per GJ than gaseous hydrogen.

The use of cryogenic hydrogen has a number of practical disadvantages, beyond those given above. Efforts are being made by BMW in FRG and elsewhere to develop

various components of the required infrastructure, such as fuel delivery tubes, quick-connects, and liquid hydrogen fuel pumps and heat exchangers in small sizes, to allow the future use of liquid hydrogen in (IC-engined) vehicles. It is however still uncertain whether the general public will be permitted to self-refuel vehicles. Boil-off of gas will occur from the tank in the vehicle, especially if it is not used for extended periods. If this is allowed to accumulate in confined spaces, such as parking garages and tunnels, it may become an explosion hazard. To prevent this, some storage device should be incorporated in the vehicle, or a catalytic recombiner should be fitted. The use of the latter will waste energy, and will result in rapid 'self-discharge' of the vehicle on stand. In contrast, a storage device might consist of a hydride or cold carbon absorber at the exit of the Dewar container. A suggested alternative would be a miniature magnetic refrigeration system, to return liquid gas (presumably catalytically converted into parahydrogen) to the container. A suitable magnetic refrigeration device does not presently exist.

In view of the disadvantages in the use of cryogenic hydrogen by the general public, one may perhaps speculate on some form of innovative storage. As we show above, a very long range can be provided by either 45 l and 45 kg of gasoline, or 3.7 kg of hydrogen stored either cryogenically in a volume of 140 l and weighing 20 kg, or absorbed on chilled high-surface-area carbon in a volume of perhaps 200 l (or less) in a system weighing 110 kg. Hydrogen could be available for delivery as cold compressed gas at temperatures close to that of liquid nitrogen, which would be inexpensive in a refueling station. If some more effective absorber than today's carbons could be identified, and if an on-board magnetic refrigeration device were available to maintain its temperature close to those of liquid nitrogen, an improved storage system might be possible. It would require a non-brittle metal inner liner to serve as a pressure container, say at 75 bar, and it would certainly require breakthroughs in absorbers and magnetic refrigeration devices. Such a system would provide about 300 W of 'cold' which could supply a portion of the cabin air-conditioning load.

Innovation is required in hydrogen storage to make the hydrogen fuel cell for a vehicle feasible. The possibility of using a fuel other than hydrogen is discussed in the following section.

7. Fossil fuel and fuel cells

When fuel cell generating plants are considered for central power plant operation or on-site use, their capital cost and lifetime must compete with competitive or semicompetitive technologies, such as natural gas turbine combined cycles. These may cost about \$800/kW today. The efficiencies of such combined cycle units are now 45%–52%, based on the lower-heating-value of natural gas fuel, but only in large sizes, for example 100 MW or more. However, they are today's most attractive technology for power production at levels about ten or more times less than central station power plants. Compared with these, they represent a dispersed generating technology option to utility planners used to conventional systems.

While gas turbine combined cycle plants are too large to be dispersed close to the points of distribution to individual buildings, they do represent an increasingly important option for utility planners, since they are available in medium-sized (100 MW or so) modular units which are small enough to be added as required to keep up with capacity demand. Their construction leadtime is relatively low, which reduces total financial exposure and interest during construction. In addition, they now have

NO_x exhaust emissions as low as 25 ppm by volume, down from 75 ppm a decade ago. This value is greater than the negligible levels of fuel cells, but much lower than those of central coal plants. The best of these, for example pulverized coal units, might have NO_x emissions of the order of 290 ppm (vol.) [10].

Fuel cells will be available in smaller units than gas turbine combined cycle plants, and they should have efficiencies on hydrogen fuel which exceed those of the latter. They should also produce zero NO_x and SO_x emissions. These factors will make them attractive provided that they can make use of inexpensive and readily available fuels. Today's society uses fossil organic fuels, and will do so for the foreseeable future. Low-temperature hydrogen fuel cells have a thermodynamic advantage over high temperature systems, but they cannot directly oxidize hydrocarbons at all. We have pointed out above that they can oxidize methanol, but at such low rates and with so many associated problems that their thermal efficiency falls from the 45–55% range (depending on the technology used) to about 30% at a system power density of only about 20 W/kg. This value is about the same as that for natural biological systems operating on carbohydrates. The low efficiency and power density of aqueous direct methanol fuel cells therefore make them uncompetitive with thermal engines. Methanol is also not a generally available fuel.

High-temperature fuel cells (molten carbonate at 650 °C and solid oxide at 1000 °C) could in principle operate directly on methanol, since its extrapolated oxidation rate would be high at their operating temperatures. However, carbon compounds tend to undergo side reactions (irreversible cracking to carbon) under these conditions. If cracking is to be prevented, a reactant to prevent carbon deposition must be simultaneously injected. This reactant is normally steam, although carbon dioxide may accomplish the same effect. If steam is used, then the methanol spontaneously reforms to a mixture of hydrogen and carbon monoxide. The hydrogen reacts at the anode, and the carbon monoxide reacts with the steam produced to give more hydrogen and carbon dioxide, until the process goes to completion or equilibrium. Thus, if methanol with steam is used at high temperatures, the fuel cell operates on hydrogen after the methanol is rapidly and spontaneously steam-reformed.

Methanol steam-reforms rapidly on suitable catalysts at low temperatures (300 °C), and it will reform spontaneously in the inlet manifolds of the high-temperature fuel cells. At 300 °C, the chemical water–gas shift equilibrium results in the reaction of most of the carbon monoxide with excess of steam to give carbon dioxide and hydrogen. In contrast, methane and polycarbon molecules such as higher hydrocarbons and higher alcohols, including ethanol, require higher catalyst bed temperatures, which are typically about 750–800 °C, where carbon monoxide is the main product. The molten carbonate fuel cells can reform natural gas with steam directly at sufficiently high rates on a suitable additional catalyst in the anode chamber in the process known as internal reforming. It will reform spontaneously without a catalyst in the higher temperature solid oxide system.

For wide use in the present fuel economy, a fuel cell system must operate on common clean fuels, particularly natural gas. Because of the constraints in reacting carbon compounds at the electrodes, all of today's fuel cell systems must first convert common hydrocarbon fuels to mixed hydrogen and carbon, invariably via steam-reforming. We should note that the reforming is a catalytic device, therefore the fuel must be low in sulfur, with a limit of about 1–2 ppm. However, this is not a disadvantage with today's clean air regulations.

The low-temperature fuel cell systems must use external reforming, with the system as heat-integrated as possible to recover waste energy, thereby increasing overall system

efficiency. All of the hydrogen cannot be used in the fuel cell, since attempting to consume anode exhaust gas with very dilute hydrogen is an exercise in diminishing returns. In consequence, spent anode gas is burned in the reformer to provide most of the heat of reaction. If the fuel cell operates at temperatures higher than the boiling point of water, its waste heat can provide reforming steam supply. Since an excess of steam is always required to complete the reaction, this would otherwise have required the burning of considerable quantities of extra fuel.

For this reason, aqueous fuel cells operating at less than the boiling point of water have not generally been considered for utility operation. These include the alkaline system and the more recent PEM systems, operating at 70 °C and about 80–90 °C, respectively. The alkaline system would in any case require complete separation of hydrogen from the gas stream, which would further decrease efficiency. The PEM cell operates at temperatures where it is susceptible to poisoning by traces of carbon monoxide in the anode fuel stream. The phosphoric acid system operates at 200 °C, which enables it to raise steam and also makes it resist the poisoning effects of up to about 1.5% of carbon monoxide. The raw gas from the reformer (largely a mixture of hydrogen, carbon monoxide and steam) is brought to equilibrium via two stages of water–gas-shift catalysts at progressively lower temperatures to reduce the residual carbon monoxide content to this level. Any further reduction in carbon monoxide content would require a more complex purification process, for example via selective remethanation.

If we consider a fuel cell operating at 0.73 V as a thermodynamic black box in which natural gas (considered here to be pure methane for simplicity) is introduced, is reformed, and is completely consumed, then the system efficiency based on the higher heating value of methane (1.154 V) will be 0.73/1.154, or 63%. The real efficiency of the phosphoric acid system operating at this voltage, including all heat losses, is about 44%. Parasitic power requirements and d.c.–a.c. conversion reduces this to 42%. The corresponding efficiency for an alkaline or PEM system operating under the same conditions would be closer to 35%. In contrast, a high-temperature fuel cell can use its waste heat not only for steam raising, but also to provide the heat of reforming. In addition, it does not require external water–gas-shift converters, which are exothermic and waste some heat. This makes it close to the thermodynamic black box, except that some fuel is wasted in the anode exhaust stream. If 90% of the fuel can be used in the cell, its gross efficiency at 0.73 V would be $(63 \times 0.9)\%$, or 56.7%.

It is clear why high-temperature plants using fuel cell waste heat for reforming should be attractive for utilities, provided that they can compete with alternative technologies in terms of capital cost. The plants are still very cumbersome, even if they contain integrated internal reforming systems, and they require complex circulations and large heat exchangers. Plants operating with integrated phosphoric acid fuel cells which require separate steam-reforming installations are at least equally large and complex, so that the whole fuel cell generation plant has a typical power density of only about 20 W/kg. In many respects, reforming fuel cell generators have many of the characteristics of large steam plants. Even a phosphoric acid cell integrated into a simple lightweight low-temperature system operating on methanol, which might allow the attainment of 50 W/kg with good design [11], is unlikely to be competitive for most transportation applications.

The high-temperature fuel cell systems can be equipped with reforming using sensible heat from the cell stack, but the large volume and low power density of today's stack technology operating on hydrocarbon or even methanol fuel makes them impossible for use for lightweight transportation applications. As we shall see below,

the high-temperature cell stacks themselves contain relatively large masses of high-temperature components, at least based on present designs. In comparison with what is required for the automobile, they are today's equivalent of Thomas Newcomen's steam engine. Like it, they are only suitable for stationary applications.

8. Utility fuel cell power densities and costs

Today's molten carbonate fuel cell stack operates at 1 kW/m^2 of cell stack active area, which weighs about 22.5 kg/m^2 , resulting in a component power density of about 45 W/kg [12]. This rather modest figure will degrade considerably when the components are assembled in complete stacks and systems, as can be illustrated by the corresponding more documented figures for phosphoric acid [13]. International Fuel Cell's large stacks for the 11 MW design operate at 0.73 V and 216 mA/cm^2 , or 1.6 W/m^2 . Cell components weigh approximately 5.4 kg/m^2 without cooling plates and electrolyte, or about 7 kg/m^2 inclusive, giving 225 W/kg on the component level. Mounting the components into 650 kW stacks degrades the power density by a factor of two, and the addition of the necessary piping and connections in pallets further degrades it by about another factor of two to about 45 W/kg . The power density of the overall system from fuel pretreatment to d.c.–a.c. conversion is 20 W/kg , about the same as that of the small TARGET program design of 1976 for a home unit rated at 12.5 kW. This weighed 500 kg in its final slim design and 725 kg in the original version [14]. The latest versions of the Westinghouse tubular solid oxide technology now operate at 0.65 V and 410 mA/cm^2 [15]. The tubular technology alone weighs about 9.5 kg/m^2 based on active area, giving the respectable power density of 225 W/kg at the tube level. In its 1986 version, the corresponding figure was 100 W/kg at a lower current density, using a smaller effective tube area. Even so, the system requires considerable quantities of ceramic materials and other components, so much so that the 3 kW demonstrator of 1986 had a net power density of only about 2 W/kg . This figure is of course not representative, but it shows that little effort has been put into the development of lightweight systems.

Typical stationary fuel cells today therefore require about 50 kg of steel and other materials per kW, which is little different from steam plants. It is therefore not surprising that their costs are high. A mass-produced 1000 kg automobile costs \$10 000, and consists of steel and other cheap materials, i.e., the equivalent of \$500 for 50 kg, or per kW of fuel cell power plant. Since fuel cell power plants are not mass-produced at present, it is not surprising that \$3500/kW is approximately the price of early model fuel cell systems. This price must be reduced in order to compete. Today, the cell stack in molten carbonate systems in Japan is estimated to cost ¥198 000/kW, or about \$1500/kW. This cost must be reduced by increasing power density, for example by a factor of two, without loss in efficiency, and by the use of lighter components and less expensive materials. An example is the substitution of copper (about \$2.00/kg) for nickel (\$7.00/kg) for the anode and anode-side cladding of the bipolar plate. This and other parts of the system require an innovative approach to design. The new technology part of the system, the fuel cell stack, requires careful attention in this respect, but so does the less glamorous state-of-technology fuel processing system, which also needs to be made lighter. Since heat transfer is the determining factor in this system, innovation is required in the design of heat transfer surfaces to increase

area per unit volume. In this respect, innovations like the flat-plate heat transfer reformer of Ishikawajima-Harima Heavy Industries might be cited [16].

The ultimate dispersed heat transfer reforming system is represented by internal reforming inside the stack itself, using stack waste heat. This is the closest approach known today to the direct hydrocarbon fuel cell system, and it requires future emphasis in the high-temperature fuel cell approaches. It also leads to simplification and weight reduction in the peripherals around the fuel cell stack, which should also lower cost. In the case of the molten carbonate fuel cell, the chemical requirement for transfer of carbon dioxide from the anode exhaust gas to the cathode also requires innovative technology. At present, the lean anode exhaust gases are simply burned. The heat produced can be used in conventional external reforming, but not in internal reforming within the stack. Unless the heat is used in co-generation, it will be wasted. Thus innovative ways of separating carbon dioxide from the waste anode stream, so that the hydrogen in it can be recycled and passed on to the cathode, are required. This will result in a much more efficient system. However the separation and transfer process must be accomplished at fuel cell operating temperatures, otherwise a complex and costly high-temperature heat exchanger system will be necessary. Elimination of heat exchangers, on the grounds of cost and reliability, to the greatest extent possible will be necessary.

New materials technology must represent a large part of these developments. Innovative new materials should allow lighter weight per unit area of active surface and higher performance per unit area. This will decrease system weight, thereby decreasing materials cost per kW. One way of doing this is the use of a coflow or counterflow configuration, rather than cross-flow, so that gas channels on one side are the indents of those on the other. This is a valid approach when metal bipolar plates are used, as in the molten carbonate system, in which consideration must be given to removal of metallic current collectors and other metal parts to lower weight, cost, and the potential for corrosion and therefore electrolyte loss. In all cases, the materials used must not be more expensive compared with those used at present, which again will require innovation and imagination. The materials must also be innovatively used, for example in radical redesigns of the solid oxide system, at the same time maintaining or improving its performance by still further reductions in the length of current pathways in each cell, to reduce IR drop. Since this will reduce materials requirements, it will reduce cost. Very thin planar structures should be the ultimate goal.

The materials must be also aimed at increasing the lifetime of fuel cell stacks from the present aim of five years to the lifetime of other plant components, typically 30 years. We should note that the design lifetime of nuclear power plants is 60 years, even though the regulatory agencies only offer licenses for 40 years. A short five-year life is particularly true for the cathode components of both the molten carbonate and the medium-temperature phosphoric acid fuel cell stacks.

In the molten carbonate cell the lithium-doped nickel oxide used at present is certainly life-limiting. While various technical fixes can extend its life, satisfactory ultra-long-term performance can only be achieved by an innovative substitute. Further innovation in the molten carbonate system is required in order to increase its power density. This is compromised today by its linear polarization behavior, which may be regarded as an excessively high internal resistance. One of the components of this is certainly internal resistance, which must be reduced. Another equally important component is the reaction resistance at both electrodes. Part of this results from electrode structures which are almost certainly far from optimization, and represent

rather what is easily attainable. Part is due to the slowness of various chemical and electrochemical steps in the reactions themselves, which are not understood. Both experimental understanding and innovation are needed here.

In the phosphoric acid system, the cell components, particularly the low density graphite bipolar plate, are already rather light. Graphite cell parts typically weigh about 5 to 6 kg/m². Because of its mechanical properties, a plate incorporating flow channels is likely to require separate structures on each side. However, as United Technologies (International Fuel Cells) was first to demonstrate, ribbing the bipolar plate is not the only way to supply a cell with reactants. This successful approach has been widely copied in Japan. Further innovation is required in both flow distribution schemes and in the manufacture of graphite repeat parts, which probably are the only reasonable possibility for stable, affordable, cell components. The phosphoric acid stack will still limit the application of this fuel cell system for on-site use if its cost cannot be lowered further. As well as the better use of materials, a higher power density would be desirable. One innovative approach is the partial (or perhaps eventually total) replacement of the phosphoric acid electrolyte by new acid materials prepared by innovative organic synthesis. Examples today are perfluorinated disulfone imide acids in gel-like (low molecular weight polymer) form, presently under examination at Clemson, Texas A&M, and Case Western Reserve Universities, and elsewhere.

Acid systems, including the PEM, suffer from a long-term problem in relying on platinum catalysts. Even if this is only used in small quantities, which will be necessarily limited by catalytic molecular turnover numbers, ultimately the application of these cells will be controlled by platinum availability. Acid fuel cells cannot therefore be considered to be a real factor in energy applications in the 21st century unless a substitute for platinum can be found. This has already proved possible at the alkaline fuel cell cathode, and it may prove easier in innovative carbon dioxide rejecting electrolyte systems operating at intermediate temperatures which use new materials and new chemistry.

When hydrogen becomes available as a fuel, the fuel cell should come into its own. Today's fuel processing systems can be eliminated, leaving at least low-temperature systems consisting largely of mass-produced cell stacks made from low cost materials operating at temperatures under 100 °C. Some of the characteristic of these stacks are discussed below. The high temperature systems will also find application, even if they are more complex, since their high-temperature waste heat will be valuable for both bottoming cycles in electric utility plants, to yield higher system efficiency, and for co-generation in on-site plants. The carbon dioxide feedback loop in the molten carbonate system is somewhat disadvantageous if hydrogen is the fuel, but with care innovative design can minimize its impact. It may even be eliminated totally if new materials or new approaches to the system chemistry can be devised.

9. Costs of vehicle fuel cell systems

An IC engine and transmission producing 500 W/kg costs \$15/kg or \$30/kW. The foregoing makes clear the fact that fuel cells will have difficulty in competing with mature internal combustion engines in regard to cost. We have pointed out above that hydrocarbon or even methanol fuel cell systems have no hope of effective use in private vehicles on weight grounds, as well as those of cost. The best phosphoric acid stacks assembled with their associated hardware today weigh 16 kg/kW, and the addition of a methanol fuel processor and its steam handling equipment is likely to

double this. The weight and volume of a 20 kW unit of this type is not possible for a 1000 kg vehicle, which would also need suitable batteries for peak power and regenerative braking.

Is a fuel cell powered vehicle therefore possible? The answer is yes, if the fuel is hydrogen, if the power system is sufficiently light, and if the materials are inexpensive, and are not in short supply. With hydrogen fuel, the vehicle will not require its own on-board refinery, just as today's IC vehicle does not operate on crude oil via its own refinery. The cost of refining crude oil to produce gasoline is about 35 ¢/gallon, or 1 ¢/kW h. Thus a refinery operating 7000 h per year costs \$70/year to operate. If the cost of capital, operation and maintenance is a typical 20% of capital cost, the refinery capital cost is about \$350/kW. Based on the electric output of a hydrogen fuel cell plant operating at 50% efficiency, the fuel processing cost will be \$700/kW, very close to that of the learned-out mass production cost of a fuel processor in a fuel cell power plant. This cost, and the weight and volume of the plant, can be eliminated from the vehicle fuel cell system if it operates on hydrogen. To be competitive with the combined cycle for utility use, where it must have a multi-year lifetime, the learned-out cost of one of today's utility fuel cell stacks should be about \$300/kW. There is no real incentive to reduce it further for this application. However, for the vehicle, the cost must come down to something in the order of \$10/kW or less.

Today's PEM fuel cell operating on hydrogen is certainly not perfect, although its performance today (3 kW/m² of active area for an atmospheric pressure H₂-air system) is excellent. It has a costly electrolyte membrane whose price (\$800–2000 per m², or \$250–600/kW for an atmospheric pressure hydrogen-air system) is not affordable. This is equivalent to \$4000–10 000/kg for a specialized plastic, which seems excessive considering the fact that the raw materials cost about \$50/kg. One would hope that mass production could lower this cost be a factor of ten, even though this will still be excessive compared with the costs of materials in an IC engine. By comparison with the electrolyte membrane, the platinum catalyst seems to be relatively inexpensive. Its present loading is a total of 0.8 mg/cm², or about 2.4 g/kW for an atmospheric pressure hydrogen-air system, based on current work at CESH. This can be reduced by 60% [17], so that its cost will then be only \$15/kW, based on a loading of 0.03 Troy oz/kW. However, 1 million vehicles per year will require 20% of world platinum production, assuming the average fuel cell only produces a modest 20 kW. Clearly, the platinum requirement will limit fuel cell application, or if all the world's vehicles in 2010 were converted to fuel cells with this catalyst loading, they would require five times the total amount of platinum which has been mined to date.

Unless some of the above can be overcome by innovation, the fuel cell power system will find use only in niche applications. Any material costing more than \$5/kW must be eliminated. The same goes for any material whose supply will limit vehicle production, so that the fuel cell can make a global impact. Basic materials used must be cheap (\$2/kg, and the weight of the fuel cell per kW must be reduced to acceptable values. As we have seen earlier, designing a fuel cell weighing 50 kg or even 20 kg per m² of active area is easy. Reducing its weight and volume further requires innovation. The Space Shuttle Orbiter fuel cell is not outstanding in this respect: its figures of merit are about 26 kg/m² for the whole system, and 14.3 kg/m² for the stack alone. Up to 1980 the complete hydrogen-oxygen systems with the best figures of merit were the early General Electric PEM Gemini fuel cell (9.1 kg/m²) of 1965, which could only operate at 37 mA/cm² on compressed hydrogen and oxygen, and the United Technologies alkaline advanced lightweight fuel cell for the US Air Force (10 kg/m²) which operated at 1 A/cm². Advanced versions of the latter

are projected to attain 2.7 kg/m^2 in the future. Today, atmospheric pressure hydrogen–air systems using lightweight components of this type could attain 3 kg/kW , or 1 kg/kW (projected). Advanced alkaline systems no longer require platinum or other noble metal catalysts, and their operating temperature of 70°C and relatively benign materials environment allows them to use, for example, polypropylene as a constructional material. Hence, $\$2/\text{kg}$ or $\$2/\text{kW}$ is attainable, again by the use of innovation.

10. Conclusions

Fuel cells are not a mature technology. If they are to make a major impact on tomorrow's energy markets, innovation in design and materials is required. Fuel cells use hydrogen fuel, so today's technology is a very ineffective design compromise to allow them to use fossil fuels. Stationary systems operating on reformed fossil fuel are expected to improve by evolutionary change, so that they will become increasingly competitive compared with, for example, gas turbine combined cycles. When hydrogen fuel is available, 60–80% of the cost of a mature fuel cell plant can be eliminated, and they should then be very competitive. However, they must not find their applications restricted by materials supply. A case in point is the platinum catalyst. Even though costs per kW can be reduced to acceptable levels, reliance on such materials will restrict fuel cell technologies to niche markets. Innovation is therefore required in materials, particularly new catalysts and new electrolytes, perhaps of the solid type, which can operate in temperature ranges which are unavailable to fuel cells today. A breakthrough in this area may even allow the direct use of carbonaceous fuels.

Fuel cells are above all an attractive technology for use in small units. These may be for the automobile, and for the broadest dispersed applications, for example, as an electrical base-load co-generation unit for homes, which would be a combined water heater and 1 kW h generator. This would operate in the first instance on natural gas using an inexpensive reformer designed for inefficiency, whose waste heat would provide water. Later, the unit would operate on hydrogen. This differs from the 1967–76 American Gas Association Target Project in that it would not attempt to be a power system to supply all peak requirements in a non-grid-connected all-gas home. The latter unit (12.5 kW) weighed 500 kg in its final form, and was optimized for efficiency, so that its cost consisted of mostly of complex heat exchangers. These would be largely eliminated in the new concept, which might weigh only 30 kg and cost $\$1000/\text{kW}$ or less. It would be supplemented for peak requirements either from the grid, or from an advanced battery. Heavy loads for air conditioning could be supplied by photovoltaic cells on the roof, if necessary. Part of fuel cell innovation will be the potential to design for new markets, which themselves will need innovation to identify.

The ultimate challenge is the fuel cell power plant for the personal automobile. The only real advantages of the fuel cell are its high efficiency (twice that of the gasoline IC engine during cruise, and perhaps three times that in urban use, with the use of regenerative braking), and its total lack of emissions at the vehicle level. Based on present technology, its disadvantages are in its materials (it may require noble-metal or other exotic catalysts) and the fact that its fuel must be hydrogen, which is difficult to store, if not to manufacture. The fuel cell stack itself must be much lighter and more compact than present units intended for stationary power applications. If it is even to be able to handle fuels such as methanol (and perhaps eventually, saturated hydrocarbons) it must use innovative materials, catalysts and internal chemistry. The most attractive candidate today would be an updated alkaline technology, based on

materials requirements. An advanced lightweight solid oxide system of monolithic construction, perhaps using a completely new family of materials, is much further away.

The future alkaline cell would use no noble metal cathode catalysts. It may eventually use polymeric amine electrolytes, whose chemistry may make them carbon dioxide tolerant. On the cell level, it must be better designed than the demonstration units available today, in which the weight of the bipolar plate may be two to three times that of active stack components, and that of the total stack is again about two to three times the cell component weight, including the bipolar plate. Lessons can be learned from the components of much lighter weight in certain aerospace stacks, since this means smaller volumes and above all, lower costs. Innovative approaches in materials and engineering design are certainly needed before the fuel cell can find applications to help reduce greenhouse gas and pollution problems in the 21st Century.

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