

GROVE ANNIVERSARY FUEL CELL SYMPOSIUM – CLOSING REMARKS

A. J. APPLEBY

Center for Electrochemical Systems and Hydrogen Research, Tees/Texas A&M University, College Station, TX 77843 (U.S.A.)

The Grove Anniversary Fuel Cell Symposium was a successful and wide-ranging meeting. It was very well attended, with a wide selection of delegates from the United Kingdom, the other European Community Countries, European Organizations, the United States, Canada and Japan. The participants included members of academia, government and industry, the latter representing both the developers and the potential users of fuel cells. It was a particular privilege to address the distinguished audience in the historic auditorium of the Royal Institution, dating from the year 1800, from the very spot where Michael Faraday and other famous nineteenth century scientists delivered their many lectures.

To begin with some remarks concerning the device itself, it should be noted that a fuel cell power generator is much more than just the fuel cell proper, *i.e.*, the d.c. power unit containing the individual stacked cells consuming hydrogen and oxygen. This part is the 'fuel cell' as assumed by the electrochemists. The d.c. power produced by this fuel cell generator must be converted to a.c., some of which is required for auxiliary systems. Water must be condensed from the fuel cell exhaust, and any spent anode gas must be used effectively. The hydrogen must be produced from primary fuel, for example methane. Using the phosphoric acid system as an illustration, the pure water reaction product will be advantageously employed for cooling the cell stack, raising steam for reforming of natural gas fuel. The endothermic heat of reforming will be provided by anode tail gas. Reformer exit gases must then be water-gas shifted to remove CO, at the same time producing more hydrogen. The plant must finally be characterized by efficient heat recovery throughout.

Overall, the plant may occupy five times the footprint or the volume of the fuel cell d.c. generator stack itself, so that the cost of the latter may only be a small portion of the total cost. Indeed, it may be only about 20% in a mature unit. The cost of the other items, which are in most cases standard chemical engineering equipment, and state-of-the-art electronic equipment, may appear to be closer to practical mass production than the fuel cell stack itself. However, the entire system today still consists of a collection of components, or rather spare parts, including a chemical engineering subsystem, quasi-integrated with the fuel cell stack, which is in turn integrated with necessary electronics, which are presently not off-the-shelf, for control and for d.c.-a.c. conversion.

Taking as an example a fuel cell system using natural gas fuel, the chemical engineering portion of the plant for the conversion of the feedstock to a hydrogen/carbon dioxide mixture has typically been a collection of parts taken from the ammonia industry. Even though this is usually considered to be state-of-the-art, it is so for a different application. A future fuel cell power generator cannot use the same ground rules as a traditional ammonia plant. It would operate under a different set of circumstances, with different economics. In a plant converting natural gas to a chemical product, such as ammonia, a different set of conditions must operate from those in a plant converting the energy available in natural gas to a different form of energy that must be economically competitive.

The very existence of an energy conversion plant will depend on its immediate economic competition. A power plant using a fuel cell is still a thermodynamic engine, with a cascade of Carnot heat-machines (pressure-temperature devices) combined with the primary electrochemical direct energy convertor. The theoretical efficiency of a thermal engine operating at a given heat sink temperature is the same as that of a fuel cell operating on the same fuel at the same temperature. This fact is intuitively implied by the second law: the maximum work that one can extract from a fuel is the free energy available in that fuel. Both devices reject a combustion product at a sink temperature, resulting in a practical work loss. The main reason why the thermal engine usually has a lower efficiency than a fuel cell is that it cannot operate at a heat source temperature that even approximates to that which can be theoretically produced by combusting the fuel. This results from materials limitations in the heat source heat exchange system. Secondly, any practical cycle has thermodynamic losses.

Similarly, the fuel cell cannot use all of the free energy available in the fuel, because of inevitable inefficiencies. These are analogous to the high temperature loss in a thermal engine. However, the high temperature thermal engine losses due to the irreversible $T\Delta S$ terms are normally greater than those in practical fuel cells. An idealized practical combination is a fuel cell combined cycle. In this, the fuel cell produces work at the thermal engine heat source temperature, so that fuel cell waste heat can be thus recovered. The high operating temperature of the fuel cell reduces irreversibly its losses, and addition of the thermal bottoming cycle compensates for the lower free energy available from the fuel at the high fuel cell operating temperature. The theoretical thermal efficiency (η) of the fuel cell alone is equal to

$$\eta = \Delta G_1 / \Delta H \quad (1)$$

where ΔG_1 is the free energy available in the fuel at the operating temperature of the fuel cell (*i.e.*, at the heat source temperature of the thermal engine, T_1) and ΔH is the heat of combustion of the fuel. A generalized formula for the maximum work available in any energy conversion device, whether it be a fuel cell operating alone, an ideal thermal engine operating at the maximum theoretical temperature (the combustion temperature of the fuel), or a fuel cell thermal engine combination, is given by the expression

$$\eta = \Delta G_2 / \Delta H \quad (2)$$

where ΔG_2 is the free energy available in the fuel at the heat sink temperature, T_2 . For an isothermal fuel cell operating alone, T_1 and T_2 are of course identical. The corresponding expression for a so-called Carnot-limited thermal engine operating between source and sink temperatures T_1 and T_2 is given by

$$\eta = (\Delta G_2 - \Delta G_1) / (\Delta H - \Delta G_1) = (T_1 - T_2) / T_1 \quad (3)$$

At the spontaneous fuel combustion temperature, $\Delta G_1 = 0$, and eqn. (3) becomes identical to eqn. (2). If a fuel cell operating at T_1 is used as a topping cycle for the thermal engine, its theoretical efficiency is given by eqn. (1), and the fraction of waste heat available for further conversion at T_1 is $(1 - \Delta G_1 / \Delta H)$. The overall theoretical efficiency of the combination is given by the sum of eqns. (1) and (3), the latter multiplied by the fraction of waste heat available. Rearrangement of this expression gives a result for the overall efficiency of the combination equal to eqn. (2). It should be pointed out that ΔG_2 in this expression is not the standard value, but that for the practical fuel conversion (utilization) desired.

Thus, if a fuel cell is used as a topping cycle in combination with a thermal engine, their theoretical losses cancel each other, and the combination behaves ideally. The practical losses (*i.e.*, irreversibilities) in a high temperature fuel cell are low, and a thermal engine can be designed to operate at typical heat source temperatures corresponding to the operating temperature of the fuel cell. The fuel cell and the thermal engine are therefore complementary devices, and a practical fuel cell 'black box'* would be such a combination. Even a low temperature fuel cell, for example phosphoric acid, benefits from a thermal bottoming cycle. A system recovering waste heat from the cells to operate a turbocompressor is an example.

The dominant theme stressed by successive authors in the Symposium revolves around the low environmental impact of fuel cells. Therein lies the political impact of the technology at the present time. Their high efficiency, though it is certainly important for the future, is less emphasized as a cost benefit in the commercialization phase than their extremely low level of tropospheric chemical and acoustic pollution. However, assuming that the fuel cells consume hydrogen, even if it is derived from fossil fuels, their high efficiency means lower carbon dioxide emissions, thus a lower greenhouse impact on global warming. These points were repeatedly stressed during the meeting. It should be noted that hydrogen used in a fuel cell produces no NO_x in the oxidation process, whereas hydrogen consumed in an internal combustion engine does. Thus, the later solution will not eliminate air pollution in cities: in fact, pollution will be almost as bad as that resulting from burning methanol in the internal combustion engine. Since hydrogen is never likely to be an inexpensive fuel, its use in the fuel cell rather than in the

*Or perhaps, due to its low environmental impact, a 'green box'.

internal combustion engine should be encouraged by the former's much greater fuel efficiency. Perhaps one can hope that future legislative credits will encourage the development of the hydrogen fuel cell automobile, because of its low social cost and because of the low capital cost for its infrastructure, which would result from its low energy requirements per kilometer.

One cannot fail to be impressed by the Japanese commitment towards fuel cell commercialization. Their industry has determined that the part of the device requiring new experience, *i.e.*, the fuel cell stack, will be manufacturable using mass production methods. It therefore will be made at costs that are a small multiplier of those for the materials alone. For the phosphoric acid system, the semi-finished materials costs for the cell stack should correspond to about \$80/kW without catalyst, and \$170/kW with catalyst. For other fuel cell technologies, *e.g.*, molten carbonate and solid oxide electrolyte systems, it is perhaps too early to tell. However, the molten carbonate system uses fundamentally simple materials technology, and should therefore not be costly when fully developed. Since the solid oxide system uses advanced ceramics with their associated difficulties in processing, it is less easy to make predictions about its future. The fundamentally simple alkaline and fluorinated acid polymer electrolyte cells, which can essentially use plastics as major construction materials, are perhaps even less defined from the viewpoint of final production cost, simply because they have been less studied from the viewpoint of the impact of the learning curve impact on their technology.

The Japanese developers and users are conducting numerous field tests of units of different technology and origin at the present time, and are thus building up a corpus of very valuable knowhow. This will give them the possibility of determining the factors influencing reliability, as well as cost, under real-world conditions. Such information, which unfortunately is not available elsewhere, either in Europe or in the United States, will place their industry in a privileged position for the manufacture of commercial units, particularly from the viewpoint of simplification and manufacturability consistent with reliability and ease of maintenance.

The continued commitment of the U.S. Agencies to fuel cell development is impressive. This has continued to be true whether they are public such as DOE, or semipublic bodies such as EPRI and GRI. Their interest in promoting the technology in the United States is still real, despite the disappointment resulting from the waning of interest by potential electric utility users. This has resulted from economic factors, particularly to the present low cost of oil-based fuel or natural gas, much less than that predicted in the early 1980s. In addition, the attitude of the electric utilities has not been aided by a perceived pricing policy on the part of developers to sell introductory units at costs beyond their real economic value, even for niche market applications. This is particularly true when the reliability and maintenance cost of early units is uncertain, and likely to be high. Thus, the cost of electricity of such units would be dominated by high capital and

(probably) O&M costs. Their improved efficiency and low environmental impact compared with competitive technologies in similar unit sizes are less important when fuel costs are low, as they are at present. However, as stated above, their good neighborliness should swing the balance in the future, away from the major competition, which is represented by advanced gas turbines and combined cycles. For on-site integrated energy systems (OS-IES) producing electricity and waste heat, which can be used for space heating and absorption-cycle air conditioning, the economic picture is brighter, since this market becomes economic at higher capital costs than those for electric utility generation. In consequence, a substantial number of U.S.-developed PC-25 200 kW OS-IES units are on order from International Fuel Cells. Some of these are being installed in Japan, along with a wide range of units developed domestically in that country.

It is encouraging that the fuel cell has again been taken up in Europe, after the pioneering work there from the nineteenth century to the late 1960s. The Netherlands has now a major program on the development of the molten carbonate high-temperature system, originally developed in that country by Broers and Ketelaar from the early 1950s onwards until about 20 years ago. Some major advances have already been made in the Netherlands in the past twelve months, notably in the development of new anode materials (with seven patent applications) and in co- or counterflow sheet-metal bipolar plate design. In Italy, a similar program is now under way, under the leadership of ENEA, with Ansaldo as prime developer. This follows the referendum held on future nuclear power in Italy in the wake of Chernobyl. No further conventional reactors will be constructed, and only power derived from advanced fail-safe reactors, or fusion systems, when these are available, will be permitted in the future. In this regard it seems a pity that there is so little interest in the development of fuel cell systems in the United Kingdom, since so many of the pioneering efforts took place here, as this Symposium has amply demonstrated. Elsewhere in Europe, the most effective effort is that on new materials, led by the EEC. This topic will be discussed later.

As stated above, the major attraction of the fuel cell power plant will be its low environmental impact. This includes lower carbon dioxide emissions compared with its rivals, especially if it is cascaded with a heat recovery cycle to obtain units with thermal efficiencies in the order of 60%, or possibly higher. When used in vehicles, it will make a major impact on the pollution problem in cities. In this respect, the United Kingdom and many European countries are 15 years behind the United States and Japan in enacting pollution controls for gasoline powered vehicles. Even these are not enough, especially in communities such as the Los Angeles basin. The situation in Mexico City, Rome and Athens is similar. Volatile reactive organic compounds, carbon dioxide and NO_x can be legislated against, but there is a limit to what can be achieved with the internal combustion (IC) engine. For example, gasoline contains enough aromatic hydrocarbons, particularly benzene, to be a cancer hazard. The fuel cell power plant can, in contrast,

eliminate these pollutants. For use in vehicles, at least based on foreseeable technology, the fuel cell power plant will require hydrogen fuel. This hydrogen might be carried on board (compressed, as liquid hydrogen, or as a hydride), or transported in the form of a hydrogen fuel carrier, particularly methanol or possibly ammonia. The use of the carrier fuel will require a load-following reactor inside the vehicle, which will be bulky and complex if high efficiency is to be achieved. An easier solution will be the use of a conversion system, probably methanol to hydrogen, in the gasoline service station or at the local level. Whether methanol will be used on a large scale as a clean-burning fuel will depend, in the United States at least, on pending clean-air legislation. In the long term, it will probably happen as oil supplies diminish and as fuel becomes based on clean coal technology. However, the oil companies are actively lobbying against methanol at present, ostensibly on safety grounds, but in practice because they favor the lower investment that will be required for reformulated gasoline. Certainly, in the short term, the 12-10 vote of the House of Representatives Subcommittee on Health and the Environment (October 11, 1989), against requiring the mass-production of alternative fueled vehicles by the automobile manufacturers, is unlikely to clarify matters.

Whether hydrogen will be available via a methanol intermediate, or as a direct energy vector, it will be necessary to store it for transportation applications. Storage of hydrogen in the vehicle is a technology that is either current, or is certainly feasible. In the F.R.G., BMW has shown that liquid hydrogen can be used, though problems associated with boil-off still remain. Even compressed hydrogen is feasible, if compressed natural gas is considered to be acceptable for an IC-engined vehicle. This can be illustrated by the increased fuel efficiency of a fuel cell vehicle compared with an IC engine, which more than compensates for the lower volumetric fuel efficiency (by a factor of three) of hydrogen in gaseous or liquid form compared with gaseous methane or liquid gasoline.

The measured urban driving cycle fuel economy of an Opel electric vehicle with regenerative braking at Texas A&M University, expressed as $\text{kW h (a.c.)}/\text{kW h}_{\text{th}}$ of gasoline is 4.1. The real vehicle power requirements at the a.c. outlet are $0.15 \text{ kW h (a.c.)}/\text{km}$. Electrical efficiency of the vehicle (ratio of a.c. input to battery output) is 57%. With an average hydrogen fuel cell voltage of 0.65 V per cell, the fuel cell efficiency will be 44% based on the higher heating value of hydrogen. Thus, the ratio of energy use as hydrogen in the fuel cell to that of gasoline in the IC engine is 1:3.17. Thus, not only will the volume of hydrogen fuel be acceptable, so will the fuel cost per km.

It was gratifying to note that Elenco in Belgium advocates the use of minibuses with alkaline fuel cells and liquid hydrogen fuel. While the fuel cell still needs improvements in materials, it most of all requires improvements in engineering. Small systems with stainless steel Swagelock components clearly require redesign, as do large systems with separate boxes joined by long segments of piping covered with hand-wrapped insulation.

The paper presented by KTI stressed this point: cost will be lowered by cascading components and eliminating separate connections. Design must be tightened up, which should be possible as reliability increases and maintenance aspects of the system, which are facilitated by modular construction, become less critical. These design aspects are being carefully addressed by the Japanese developers in their field test programs. For stationary systems, weight is at first sight not very critical. For example, in the phosphoric acid system, the weight of active stack components (minus electrolyte) in the Westinghouse unit is now about 4.5 kg/m^2 , 90% of which is the cross-flow graphite bipolar plate. This represents 3.75 kg/kW at current power densities. Thus, the active components represent about 1500 kg per stack. However, the weight of the latter, without the pressure vessel, is about 7.2 tonnes. Since weight is synonymous with cost, it would seem that lightweight designs must be evaluated in the future. In fact, the impression is that today's fuel cell designs are largely breadboards, made for accessibility rather than for optimum cost. The stationary systems (phosphoric acid, molten carbonate, solid oxide) all require simplification, lower weight, and thus lower cost. As an exaggerated example, the 3 kW solid oxide units contain approximately 25 kg of tubular cells, about 80% of which is the weight of the support tubes. However, the weight of the total system is 1300 kg. The phosphoric acid system still requires performance improvements to compete with the combined cycle. In contrast, the molten carbonate system can reach much higher efficiencies than this competitive system, but it still requires some materials improvements, for example, at the cathode. It also requires a cost-effective bipolar plate with cost-effective aluminizing for corrosion prevention. Similarly, the solid oxide system requires weight reduction and cost-effective manufacture.

For mobile power fuel cell systems, cost must be greatly reduced to levels that are perhaps 10% of those for stationary applications. Specific power must also be increased. Again, both of these imply weight reduction of the repeat parts of the cell stack. For a PEM system operating on air at ambient pressure, a maximum current density of about 4 kA/m^2 can now be obtained at 0.6 V. This represents 2.4 kW/m^2 , or 480 kW/m^3 at a standard stacking pitch of 5 mm for cross-flow cells of standard configuration. Using present 'standard' components, this corresponds to 1000 kg/m^2 , or 2.1 kg/kW for the basic d.c. module repeat components. However, it should be possible to improve the engineering design still further by improving the engineering design of the stack. Typically, a large graphite bipolar plate intended for a utility phosphoric acid system has 1.5 mm square gas distribution grooves in a cross-flow configuration in a plate with 0.5 mm web. A smaller plate for a mobile PEM system may have grooves only 1 mm square, allowing a stack specific weight reduction of about 20%, to give 4.1 kg/m^3 and 1.7 kg/kW , at the increased specific power per unit volume of 600 kW/m^3 , resulting from the increased stacking density (4 mm cell pitch). If internal manifolding and a co- or counter-flow arrangement are used, the structure of the bipolar plate can be made much lighter and thinner. In such

a case, the grooves are parallel on each side, and if the pitch on one side is displaced 90° out-of-phase with regard to the other, a thin folded or undulated plate can supply the gas channels, as in the Alsthom design of the late 1960s and early 1970s. As in this bipolar plate, a graphite-plastic composite may be suitable, though other possibilities exist. If this should be the case, a plate weighing 1.5 kg/m^2 is possible, giving a total component weight of 1.8 kg/m^2 , including electrodes, electrolyte layer, and a wicking arrangement for water mass control, for example, by the use of thin conducting graphite felts. A cell pitch of 2 mm should therefore be possible, allowing a power density per unit volume of 1200 kW/m^3 , with a specific weight of 0.75 kg/kW .

Attempting to achieve this goal has nothing to do with electrochemistry, and everything to do with good engineering design. Electrochemically speaking, it is based on the state-of-the art current densities and power densities for the latest PEM cells operating under atmospheric pressure conditions at Los Alamos National Laboratory and at Texas A&M University. Naturally, such ambitious performance will require innovative engineering design in other associated components and subsystems, including cooling and gas flow distribution and control with allowable geometry for pressure drop. It also requires innovative lightweight end-plate design, with elimination of the standard tie-bolts and their replacement by, for example, a sealed tension system incorporating composite structures. For the ambient pressure PEM system, it is perhaps possible to achieve 1.125 kg/kW for the complete stack, with its hydrogen fuel and air supply and cooling systems.

These energy densities may seem to be unrealistic, until one considers that a projected alkaline fuel cell (International Fuel Cells) that will operate at 80 kA/m^2 on pure hydrogen and oxygen at 13.6 atm is now a real possibility. The projected weight for this system per unit area is 1.8 kg/m^2 . For the cell stack alone, including an advanced cooling system and its auxiliaries and the pressure vessel, 7 kW/kg is projected. For a complete system, physically similar to that in the space shuttle, 300 kW is expected in a 90 kg unit. Such power densities are of the same order as those from military gas turbines, and they largely exceed the possibilities of normal internal combustion engines. To put them in another more historical perspective, the mid-60s alkaline fuel cell used in the Apollo program weighed 115 kg and produced 1.5 kW , whereas the three-stack version of the mid-70s space shuttle orbiter system produced 18 kW (limited by cooling) for the same system weight. Both of the above operated on cryogenic hydrogen and oxygen at 4 atm pressure.

A 'terrestrialized' atmospheric pressure version of this generator, operating on hydrogen and CO_2 -scrubbed air at 70°C (rather than 150°C) could use inexpensive materials (plastics) and non-noble catalysts (at least at the cathode). With a pyrolyzed cobalt TAA-carbon catalyst (or similar compound), 4.5 kA/cm^2 at 0.65 V should be achievable, giving a power output of 60 kW from a stack weighing 40 kg , occupying 41 l . The complete system might weigh 90 kg , so that its weight and volume will be compatible with a

family car. In mass production, an alkaline system could be inexpensive (perhaps only a few dollars per kW for the stack, based on an average materials cost of \$2 - 3/kg or \$4 - 6/kW, of which only 5% would be cobalt).

Similarly, there is no reason why a PEM should be expensive in the future. The present cost of Nafion[®] is over \$3000/kg. It is made from the same starting material (tetrafluoroethylene) as Teflon[®], which costs about \$40/kg. The differential is disproportionate, but the cost of Nafion[®] is artificially inflated by its value to the chloro-alkali industry. There is no real reason why a PEM fuel cell stack, 10% of whose weight would be fluorocarbon acid polymer, could not also have an average materials cost that would ultimately not be very different from that of an alkaline stack. Even platinum electrocatalyst cost can be (marginally) acceptable: state-of-the art loadings of 1 and 2 g/m² can now be achieved at the anode and cathode respectively, based on work at Los Alamos National Laboratory and Texas A&M University. This represents 2.4 troy oz (75 g) for a 60 kW unit, *i.e.*, \$22/kW. However, unless a major breakthrough in acid-electrolyte catalysis takes place, the use of platinum catalyst will be impractical: at the present catalyst loading, only 1.3 million cars per year can be supplied from the entire world production of platinum*. The present world vehicle population, with a 10-year turnover time, is 500 million, and it is projected to be 2 billion by the year 2010. Thus, a platinum-based fuel cell system could not begin to make an impact on the energy use of future vehicles and on their exhaust emissions.

Based on present knowledge, the best candidates for mobile applications will be the alkaline system and, more problematically, a monolithic solid oxide system, if it can be made. The difficulties involved in achieving this goal were carefully and convincingly summarized at this Symposium in the paper from Combustion Engineering. A definite advantage of the solid oxide cell is that its waste heat can be used in a thermal bottoming cycle, *e.g.*, by pressurizing the system, which will allow an increase in power density and perhaps in efficiency, thus further reducing cost. The power density of the low temperature systems will benefit greatly by pressurizing, but then extra electrical energy must be used to supply the high pressure air, unless some innovative method of compression can be developed, for example using the energy available in liquid hydrogen. For example, if this is stored under 70 atm pressure, it could pressurize ten times its volume of air (a typical requirement) when let down to 5 atm. While many authors suggest the use of pressurized PEM cells, the electrical requirement to provide pressurization may be the equivalent of 0.15 V, which would

*Editor's note: although a fuel cell powered car will not require a precious metal catalytic exhaust convertor; hence this application represents a switch in the location of platinum catalyst rather than a wholly new requirement. This aspect of supply will need to be confronted in either event, although fuel cell platinum catalyst levels (where required) would be two orders of magnitude higher than those for catalytic mufflers based on state-of-technology loadings.

seriously degrade overall system efficiency if it had to be provided electrically.

For the future, hydrogen must be given pre-eminence as the fuel cell fuel par excellence. While methanol may be the preferred future fuel for vehicles, this does not necessarily mean that a fuel cell vehicle should carry around a bulky load-following chemical plant. As already noted, a better solution would probably be to place a reformer at the service station. To 'fill the tank' of a hydrogen fuel cell car might typically require $150 \text{ kW h}_{\text{th}}$ of hydrogen to give 450 km range. A gasoline pump can service about 10 vehicles per hour, 12 hours per day. Thus, a typical six-pump station would require a 4.5 MW reformer, operating continuously with hydrogen storage. This does not seem to be an impossible requirement.

Finally, the key to the future is the development of new materials. Not everything has been invented yet: new materials may allow a broadening of the scope of fuel cells, and allow their wider use. New solid electrolytes are part of this opportunity, and it is gratifying to know that the EEC has a far-sighted program in this area. The risk is high, but the opportunities and potential spin-off are great, for example, in the field of catalysis and superconductivity. New lines for research exist in the area of new materials, which will serve to educate the electrochemists and electrochemical engineers which will be required for the future world of the electrochemical engine.