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Status of solid polymer fuel cell system development

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Abstract

Solid polymer fuel cell (SPFC) systems are expected to see service in a wide variety of applications, including road vehicles, trains, ships, undersea power, and small scale stationary power generation. Each application brings unique requirements in terms of fuel, power, efficiency, volume and weight and, consequently, SPFC systems are expected to take a variety of forms. This paper reviews the development issues which must be resolved before SPFC systems can enter commercial service. It includes the results of system studies completed by Rolls–Royce and Associates during the last two years. Development priorities are highlighted, particularly for the stack and fuel processing system. Results of the testing of a novel compact fuel processing system are presented.

Introduction

Solid polymer fuel cells (SPFCs) are potentially a highly efficient means of generating electricity. However, they are only of practical use as part of a complete power generation system, converting a viable fuel into useful grid quality electricity, or power transmission for a vehicle. The ultimate commercialisation of SPFCs will therefore demand that this system package is optimised to translate the SPFC potential into the requirements for a given application. This will necessarily result in performance and cost trade-offs over the various components, including the SPFC stack. Appreciation of the system requirements, system integration issues and relative consequences, and the ultimate commercial targets are therefore very important.

With just these points in mind Rolls–Royce and Associates (RRA) have recently complete a study as part of the UK Department of Trade and Industry's Advanced Fuel Cell programme, to help identify the key R&D issues with the SPFC system [1].

The variety of potential applications place different constraints and targets for the system design. The inherent qualities of SPFCs (high power density, high full and part-load efficiencies, low temperature, responsiveness, robustness) make it an attractive proposition for transport, but it also has potential for power generation, particularly for stand-alone application or cogeneration.

The features required for this range of applications are indicated in Table 1. The variety clearly illustrates that different stack and system designs will be required for each and different integration issues must be resolved.

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TABLE 1

SPFC system targets for potential applications: approximate system targets

Application	Likely SPFC power range	Cost ($)	Efficiency (%HHV)	Operating mode	Fuel choices	Key design constraints (excluding cost)	Competing technologies
Stationary stand-alone	<50 kWe	<1500/kWe	>30	Load follow	Nat. gas LPG	Efficiency Transient response	Diesel generators
Cogeneration grid connected	<200 kWe	<750–1200/kWe installed	>36 Elect >75 Total	Slow load follow	Nat. gas	Efficiency Heat recovery	Other fuel cells Gas ICEs
Submarine Battery Hybrid	>100 kWe	<15000/kWe	>40	Steady state	Methanol Hydrogen LPG Kerosene Diesel	Fuel/oxidant storage Compactness Efficiency	Alkaline fuel cell Closed cycle Heat engines
Service vehicle Battery Hybrid	~100 kWe	<180/kWe	>30 average	Slow load follow	Methanol CNG LPG Hydrogen Kerosene	Fuel storage Start-up	Diesel ICEs
Electric vehicle Car	~50 kWe	<90/kWe	>25 average	Rapid load follow	Methanol CNG LPG Hydrogen Kerosene	Start-up (1 min) Transient response Compactness Fuel storage	Petrol ICEs Battery

System design

Design features

The incorporation of appropriate features for a viable design can result in a complex arrangement, as is illustrated in Fig. 1. This shows a process diagram for a 50 kWe cogeneration system, using natural gas as the fuel and incorporating a turbocompressor for air delivery. This 'simple' arrangement is not considered to be optimised, but provides a good illustration of the complexity and some of the integration issues which must be resolved in a successful design. These include:

(i) *Choice of fuel.* This must be commercially viable, readily available, easily transformed into hydrogen and, in the case of transport applications, have compact storage characteristics. There is as yet no fuel technology which meets all these requirements.

(ii) *Choice of reforming technology.* Well-established industrial reforming technology must be scaled down to sizes appropriate for the majority of SPFC applications (<200 kWe). In addition, improved start-up, responsiveness, compactness and system integration are all required to meet the various application requirements.
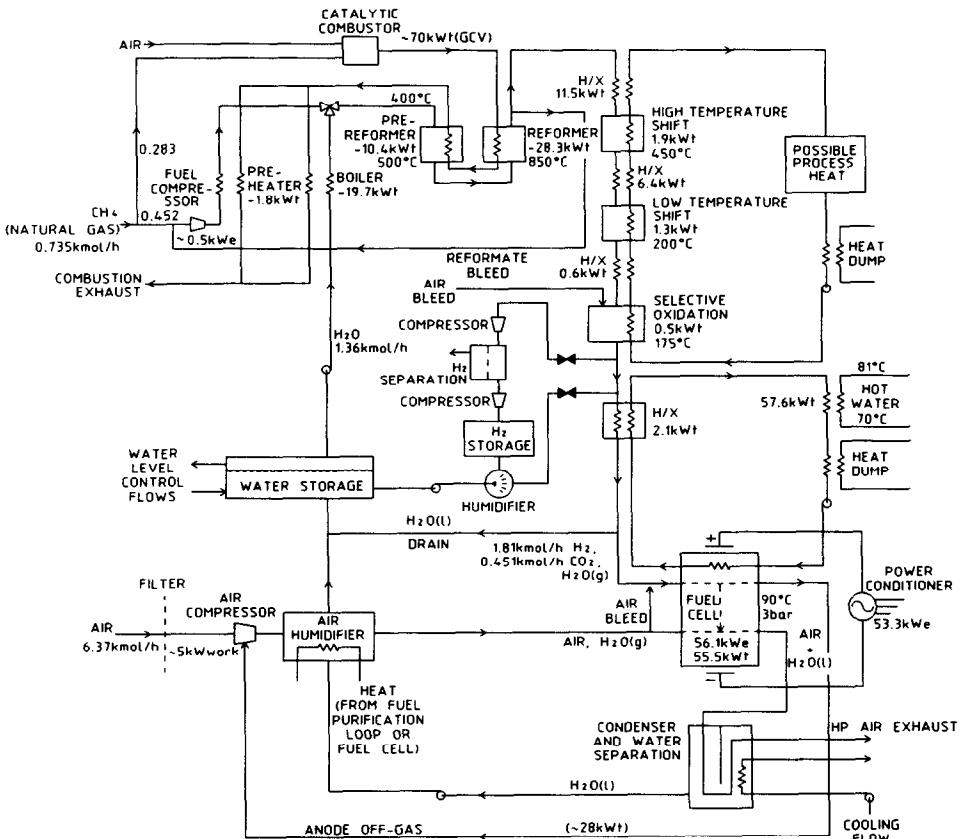

Fig. 1. Example of process requirements for SPFC system for cogeneration application.

(iii) *Choice of reformate purification* (removal/reduction of carbon monoxide/sulfur, etc.). The range of options (selective oxidation, separation techniques, etc.) all impact the overall system operating conditions, size and cost.

(iv) *Optimisation of operating pressures/temperatures/reactant flow rates*. The balance between component and system performance and cost is a key feature of a successful system design.

(v) *Use of anode off-gas*. With the exception of pure hydrogen, continued recirculation of the anode off-gas will not be possible in a full system. The retained energy of the anode off-gas (typically ~20% of input fuel energy) must be used effectively. This will almost inevitably result in some time-lag in the system, requiring careful design and control to maintain a suitable transient response.

These and many other issues must be resolved in a successful design. The correct balance must be maintained between performance and cost of all components to ensure a well-integrated and ultimately commercially viable system.

Typical factors involved in this balance are indicated by Tables 2 and 3 and Fig. 2, both of which are drawn from the results given in ref. 1. Tables 2 and 3 show some of the relative differences in performance and system size for a variety of arrangements for a cogeneration system. Similar results would be applicable over the range of potential applications.

The impact of varying fuel, operating conditions and system integrations are readily apparent. It is also obvious that the nett efficiency is considerably lower than the stack efficiency, which must be kept high to compete with alternative technologies.

Cost considerations

Figure 2 provides an illustration of potential system cost splits, based on current and potential future SPFC system technologies. (Note that the cost estimates ignore recovery of design and development costs, but are based on current and projected manufacturing techniques, material costs, and performance parameters taking account only of technology advances which may be anticipated today, e.g., transfer of existing single-cell results to a full stack.)

Currently stack costs dominate, showing that significant advances are required in reduced stack material costs, using cheaper alternatives or reducing quantities of expensive components. Equally important is the development of manufacturing techniques, to remove labour-intensive processes and improve the quality of production. This will require changes in stack design from those currently developed from laboratory experiments. Note that even taking into account the improvements currently noted in laboratory tests (low platinum loading, lower cost membranes, high power density), stack and system cost projections are only just approaching acceptable levels for the potential high cost applications. This clearly indicates that there are still some significant progress to be made in stack design, which will potentially demand radical redesign (for example, stack voltages of ~600 V are desirable for systems integration, compared with current voltages ~50 V).

Another factor illustrated by the future cost projections is the far more even distribution of costs around the whole system. That is, it can be foreseen that SPFC stack developments already underway will reduce costs to the extent that the stack no longer dominates the system package. It is therefore equally important to develop other system features to achieve a competitive design, such as the fuel processor, air delivery and power conditioner. The development of such well-established technologies as air compression systems and hydrocarbon reforming will also demand significant

TABLE 2

50 kWe natural gas cogeneration system options (using steam reformers)

Option description	Option number	System[a] efficiencies		Relative size[b]			
		Electrical	Stack	Fuel processor		Fuel purification	Total air flow[c]
				Reformer gas flow	Combustion gas flow		
As Fig. 1, with low (92%) fuel conversion in reformer	SR-1	26.1	1.0	1.0	1.81	1.0	(1.0)
As Fig. 1	SR-2	27.5	1.0	0.98	1.81	0.98	(1.0)
As SR-1, reformer temperature +600 °C	SR-4	23.1	1.0	1.32	1.73	1.04	(1.0)
As SR-1, electrical air compressor, off-gas combustion to help heat reformer	SR-5	27.7	1.17	1.17	2.34	1.17	1.17
As SR-5, pressure recovery from exhaust air	SR-7	29.7	1.09	1.09	2.19	1.09	1.09
As SR-5, 60% increase in air flow	SR-8	25.7	1.29	1.29	2.45	1.29	2.02
As SR-5, 95% hydrogen utilisation	SR-9	29.1	1.39	0.99	1.99	0.99	1.17
As SR-5, hydrogen humidification using stack heat rather than excess steam in reformer	SR-10	29.7	1.17	0.83	2.04	0.96	1.17
As SR-5, cell efficiency +61% (0.9 V)	SR-11	34.5	0.94	0.94	1.88	0.94	0.94
As SR-5, system pressure +5 bar	SR-12	27.9	1.20	1.14	2.26	1.19	1.20
As SR-5, stack temperature +80 °C	SR-13	26.8	1.25	1.18	2.35	1.23	1.25

[a]Efficiencies are related to the HHV of total fuel input. Total efficiences include heat recovery for cogeneration, with the range covering possible air humidification requirements (100->0%).

[b]Relative sizes are as referred to SR-1.

[c]Values of total air flow are provided, at least in part, by a turbocompressor.

TABLE 3

50 kWe natural gas powered cogeneration systems — other reforming options

Option description	Option number	System[a] efficiencies		Relative size[b]			
		Electrical	Stack	Fuel processor		Fuel purification	Total air flow[c]
				Reformer gas flow	Combustion gas flow		
As Fig. 1, with low (92%) fuel conversion in reformer	SR-1	16.1	1.0	1.0	1.81	1.0	(1.0)
As SR-1, with autothermal integrated heat transfer between purification/preheat, electrically-driven compression of ATR air supply	ATR-1	27.7	1.08	2.36		1.96	(1.42)
As ATR-1, 100% fuel conversion in reformer	ATR-2	28.9	1.08	2.34		1.96	(1.42)
As ATR-2, high-efficiency electrically-driven air compressor	ATR-3	29.3	1.06	2.31		1.92	(1.40)
As ATR-3, high-efficiency electrically-driven air compression	ATR-5	31.0	1.05	2.12		1.76	(1.34)
As SR-1, with partial oxidation reforming of methane	POX-1	19.0	1.15	3.01		2.45	(1.95)
As SR-1, with hydrogen separation from reformate using membranes (includes high pressure reforming)	HSR-1	26.6	1.05	1.32	2.60		(1.05)
As SR-1, with methanol fuel, reformed at 300 °C	MSR-1	31.2	0.99	0.92	1.0	1.04	(0.99)

| As SR-1, with hydrogen fuel, supplied direct to the stack | H-1 | 34.9 | 1.03 | (1.03) |
| Maximum realistic potential performance from a system using hydrogen fuel, assuming 10% system parasitics and cell voltage = 0.9 V | H-4 | 53.4 | 0.84 | 0.84 |

[a]Efficiencies are related to the HHV of total fuel input. Total efficiencies include heat recovery for cogeneration, with the range covering possible air humidification requirements (100–>0%).
[b]Relative sizes are as referred to SR-1.
[c]Values of total air flow are provided, at least in part, by a turbocompressor.
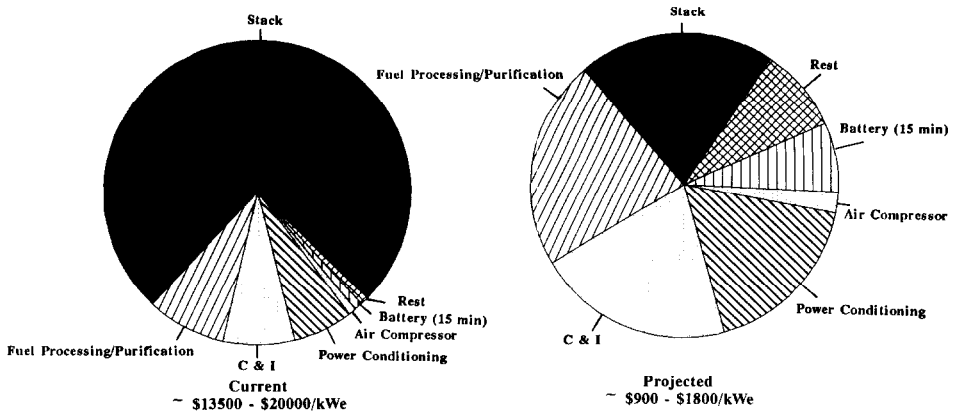
Fig. 2. Indicative current and projected system costs, based on 50 kWe cogeneration system.

effort and some innovative approaches. These must be progressed in parallel with the stack to ensure that a commercially viable system results.

Summary of system development issues

From considerations such as those illustrated above, it is clear that advances are required across the full technology spectrum to take SPFC systems successfully to the market place. The key areas of cost, performance and integrity have yet to be fully addressed and must be tackled to provide an attractive product for the potential customer.

Specific issues includes:

(i) reduced material costs;

(ii) manufacturability of stack and component designs;

(iii) fuel processor start-up times and system responsiveness;

(iv) reduction of parasitic loads, particularly air compression;

(v) development of suitable 'low cost' purification systems;

(vi) broadening of stack performance envelope, including poison tolerance, humidification and pressure, and

(vii) optimum system integration.

These issues have particular significance for the design of the fuel cell stack (page 136) and the reformer (page 137).

Fuel cell stack

The cost targets for a commercial SPFC stack vary with application. In stationary applications the cost must be reduced to below US\$ 200/kW, whereas for vehicle applications cost levels of US\$ 30–100/kW are required, depending on the system and duty.

The performance targets for a commercial system will also demand some improvement in the current state-of-the-art, particularly in the following areas:

(i) *Volume.* The available volume varies with application. Typical vehicle applications will demand that the stacks including humidification, recirculation, cooling, and any stack interconnections are packed into a volume of 1 to 2 l/kW.

(ii) *Output voltage*. Electrical connection and conditioning costs are high if the stack output voltage is too low. In most applications the stack output voltage must be raised into the 200–600 V region.

(iii) *Air delivery conditions*. Current stacks are often operated with significant air oversupply (over 1.5 times stoichiometric) and at over 2 bar. These requirements can result in a parasitic load of up to 20 to 30% of fuel cell output. For a given net power output they also increase the cost and size of the stack and the supporting systems.

(iv) *Poison tolerance*. If reformate is used poisoning becomes an important consideration. While there are many potential methods of removing poisons they tend to impact system cost, volume, integrity or start-up time. A more poison tolerant stack would ease these concerns.

(iv) *Stack power*. In most applications a single stack with a power tailored for the duty will be required to simplify installation. Existing stacks generally have rather small power ratings.

Taken together, the cost and performance targets for a commercial SPFC stack are challenging. It is not possible, at this stage, to plan or cost a development programme which will meet these targets. Some aspects of such a programme are dependent on the progress of research which is by its nature unpredictable. Likewise, the introduction into service of SPFC systems will be dependent on legislative factors, environmental policies, fuel availability and other factors which are difficult to predict. Given these risks, it is doubtful whether any manufacturing organisation could commit at present to the substantial development programme and investment in production facilities which must precede the manufacture of a commercial stack. RRA's approach has been to conduct a modest investigative programme which has included:

(i) studies of novel SPFC stack concepts;

(ii) construction of substack units using novel manufacturing techniques;

(iii) tests of single cells (100 mm × 100 mm) with the aim of understanding stack and system design issues;

(iv) purchase and testing of a stack incorporating novel manufacturing techniques.

To date, we have formed the view that two aspects must dominate a commercial stack development programme:

(i) *Stack mechanical design*. A future commercial SPFC stack will probably look somewhat different internally to traditional units. Detailed material and manufacturing considerations for nonactive parts of the stack — bipolar plates, end plates, manifolds, humidification, and cooling — will ultimately dictate the preferred design. Innovative thinking is required, and the skills of the system designer, production engineer and material scientist must by brought to bear on the problem.

(ii) *Fuel cell process/thermal design tools*. The commercial target is a practical full size fuel cell with low catalyst loading which will operate on reformate/air at modest pressures and without excessive fuel/oxidant oversupply. Performance requirements lead towards high operating temperature with minimum humidification and thin electrolyte membranes. The design of such a system requires a sophisticated understanding of the processes occurring as the reactants pass through the cell.

Overview of reformer types and effects on overall system efficiency

Reforming

Reforming is a term that refers to a collection of techniques for processing hydrogen-containing chemicals to produce a hydrogen-rich gas (reformate). Although

the three major techniques described below may be carried out either catalytically or noncatalytically, the catalytic processes offer better control over product selectivity and allow for smaller plant. Further discussion of reforming will therefore be concerned only with catalytic processes.

Steam reforming

Steam reforming, in which water is used as an oxidant to liberate hydrogen from the hydrocarbon, is the most established reforming technique. All steam reforming reactions are highly endothermic requiring a significant input of heat from an external source.

The requirements for an external steam supply, extensive heat-transfer surfaces and a separate combustion arrangement tend to make steam reformers bulky and slow to start.

Partial oxidation reforming

The partial oxidation technique uses oxygen (or air) as an oxidant. Partial oxidation is highly exothermic and hence self-sustaining. Consequently, partial oxidation can be carried out in simple packed bed reactors. Partial oxidation produces only two moles of hydrogen per mole of methane as opposed to four moles by steam reforming. Unless provision is made to recover the waste heat, partial oxidation is less thermally efficient than steam reforming.

In addition the hydrogen content of reformate from a partial oxidation reformer is lower than that from steam reformer. If air is used as an oxidant rather than oxygen then the proportion of hydrogen is further diluted by the nitrogen content of the air.

The exothermic nature of the reaction is such that start-up and load following can be extremely good.

Autothermal reforming

Autothermal reforming is a term used to describe a hybrid process which uses a mixture of steam and oxygen (or air) as an oxidant. The heat liberated from the exothermic partial oxidation reaction being used to drive the endothermic steam-reforming reaction.

Autothermal reactors have the potential for combining the rapid start-up and load following capabilities of the partial oxidation reformer with the efficiency of the steam reformer.

Carbon monoxide formation and removal

Carbon monoxide formation is of interest, because it acts as a severe poison towards the SPFC anode catalyst in concentrations greater than a few ppm.

The concentration of CO in the reformate leaving any of the reformers described previously is governed by the water-gas-shift reaction:

$$CO + H_2O = CO_2 + H_2 \tag{1}$$

Under equilibrium conditions the left-hand side is favoured at high temperatures whilst the right hand side is favoured at low temperatures. As a result of this reaction the synthesis gas produced by reforming reactions (always carried out at high temperatures) contains a significant ($\approx 8\%$) proportion of CO.

Carbon monoxide levels can be reduced by allowing the reformer product gases to cool before passing them over a second catalyst (active for the shift reaction only) at lower temperatures when the shift equilibrium is to the right, thereby reacting CO with steam to form CO_2 and H_2.

Due to equilibrium and catalyst operating temperature limitations it is not practical to reduce CO levels below about 2000 ppm by this method.

Importance of new reformer options for SPFC integration

The choice of the best reforming technique for any particular application is not simple. What is certain is that steam reforming, which has become a fairly automatic choice for phosphoric acid fuel cells (PAFCs) is not necessarily the best choice for SPFCs. The bulky, slow starting steam reformer degrades the most desirable attributes of the SPFC. In addition, an SPFC (unlike a PAFC) does not provide a source of waste steam.

Autothermal reforming has some disadvantages compared with steam reforming, particularly the additional N_2 and CO_2 in the product stream. This results in a small increase in pumping power, and increases the size of the shift reactors. It is also important to ensure that the stack is designed to accommodate this nonreactant gas without the anode off-gas having an excessive H_2 content.

These disadvantages need to be balanced against the advantages of autothermal reforming. In a properly integrated autothermal system the thermal efficiency can be higher than a steam reformer because there is no energy loss in the flue gas. This efficiency gain more than compensates for the increased pumping power mentioned above (see Tables 2 and 3). Furthermore, the autothermal operating point can be adjusted to be exothermic (e.g., if additional heat is required in a vehicle or cogeneration role) or endothermic (e.g., if it is desired to utilise heat from anode off-gas).

Much emphasis is currently being placed on methanol as a fuel for fuel cell transport applications. Methanol has a number of advantages, primarily because it is a liquid at normal temperature and pressure and because it is very easily reformed at low temperatures (~ 250–300 °C). Its disadvantages lie in its toxicity and the current lack of any distribution infrastructure. The conditions under which methanol is typically reformed can also lead to the reformate gas containing poisons such as formic acid and formaldehyde which require cleaning before the product can be used in a polymer electrolyte fuel cell. All other common hydrocarbon fuels including ethanol require the use of high temperatures to liberate hydrogen.

A high-temperature autothermal reforming system has the significant advantage that with little modification a variety of fuels ranging from natural gas to kerosene could potentially be utilised in the same unit. Modifications to accommodate different fuels would be concerned with provision to vaporise liquid fuels, provision to change catalyst systems to accommodate heavier hydrocarbons which may be prone to coke deposition and the ability to remove sulfur at varying concentrations. A high-temperature system could also be configured to operate on methanol. Such a system would have the further benefit of being equally applicable in either a transport or stationary application.

The inherent attractions of a high-temperature autothermal reformer (integrated design, high efficiency, fast response, multifuel capability and ease of construction) are clear benefits for a commercial SPFC system.

RRA autothermal reformer performance

During 1992 RRA commissioned a small autothermal reformer system. The unit is experimental, i.e., designed to permit modification and development of the process stages. The unit is a single vessel which takes its supplies of water, methane and air at ambient temperature. If differs from a practical unit in that it does not incorporate

desulfurisation or a selective oxidiser for final CO removal (although the vessel contains space for these facilities). It operates at ambient pressure to ease modification, but the processes could operate equally effectively if pressurised. The total volume of the vessel is 60 l.

Tests have investigated the full range of operating conditions studying the effects of air:methane ratio, steam:methane ratio and methane throughput on the performance of the catalyst systems.

Figure 3 shows the efficiency of the unit over a range of input power levels from 7.5 to 29 kW (equivalent to approximately 2.5 to 10 kW of electrical output). The thermal efficiency, calculated by comparing the higher heating value (HHV) of input methane and product H_2, is well above 90% through most of the range of operation.

Figure 4 shows the carbon monoxide concentration in the product gases at a range of power levels. These show that the CO concentration is below 2100 vpm throughout the power range.
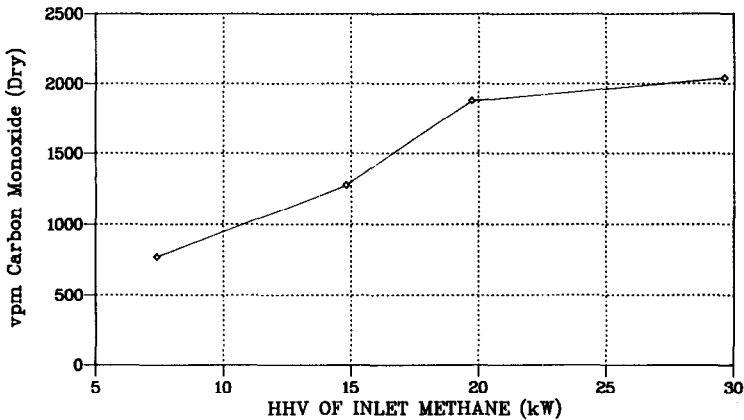

Fig. 3. Rolls–Royce and Associates' autothermal fuel processor outlet CO concentration vs. methane throughput.
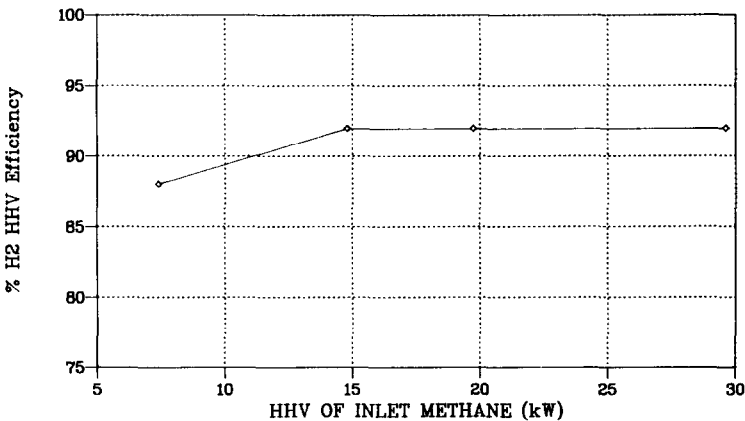

Fig. 4. Rolls–Royce and Associates' autothermal fuel processor H_2 generation efficiency vs. methane throughput.

Future SPFC reformer designs

RRA envisage that SPFC reformers will take a variety of forms depending on the application. We foresee that autothermal reformer units could be developed with a range of characteristics to match the majority of the system requirements listed in Table 1.

Effort is now being directed towards improving system start-up time. The key issue here is not the heating of the reforming bed — which is relatively rapid — but bringing downstream CO clean-up stages rapidly onto condition. Desulfurisation is also an important question during start-up. Our target is to achieved a twenty-minute start-up time for a natural gas powered system including reactors and desulfurisation by mid 1994.

A view of potential early SPFC applications

The SPFC is the most flexible of all fuel cell types. Whether and when it achieves commercialisation will depend principally on how far and how quickly the cost of the stack can be reduced. Assuming that acceptable costs can be achieved, what are the possible early applications of SPFCs, and what will be the important features of such systems?

Transport applications

SPFCs have very special characteristics for transport duties. There are few alternatives to the SPFC if a pollution-free vehicle with adequate performance and range is desired. In the longer term the combination of the SPFC with lightweight hydrogen storage appears attractive. In the shorter term it will be necessary to operate the SPFC with a practical hydrocarbon fuel and a fast starting compact reformer system. Start-up and transient energy storage will be key issues since both hydrogen and electricity tend to be heavy and expensive to store. There is a strong incentive to minimise this storage problem by improving the start-up characteristics and transient response of the fuel cell system and in particular the reformer.

The choice of hydrocarbon fuel for transport systems has been the topic of much debate. The issue will eventually be settled by a combination of cost constraints, government policies and customer preferences, and the outcome is therefore difficult to predict. It will be wise to develop SPFC system technologies which do not stand or fall depending on the outcome of the fuel debate: i.e., they have fuel flexibility.

Stationary applications

In contrast to transport systems, stationary applications of SPFCs are superficially less attractive. The steady operating efficiency of a hydrocarbon fuelled SPFC system will be 25 to 38%, and heat recovery from the system is difficult. There is a natural tendency to compare SPFC systems with existing gas-powered cogeneration systems, and on that basis the case for developing an SPFC-based cogeneration system appears marginal. However, these comparisons, although useful, can oversimplify the situation and ignore some key facts:

(i) Many small stationary power systems are not grid connected. Even when a grid is available the price of maintaining it to meet peak demands only may be high. Grid independent units must be sized to cope with peak loads but operated for long periods at part load. Under these circumstances the efficiency of a gas engine can fall whereas the fuel cell offers potential for good part load efficiency and has short-term overload potential at reduced efficiency.

(ii) The noise and emissions from a gas engine can be suppressed but only at the price of additional installation costs.

(iii) Maintenance costs of a gas engine can be high, particularly in remote locations.

(iv) Good transient response is important for grid independent operation, giving the SPFC an advantage over other fuel cell types.

For these reasons we would take the view that the SPFC is attractive for grid-independent power duties provided that the initial cost is competitive. Following its introduction in these applications its use may spread into cogeneration.

Conclusions

The SPFC has great potential to replace internal combustion engines (ICEs) in small-scale applications, particularly those where operational flexibility and compactness are important. If the SPFC is widely introduced the resulting benefits in terms of reduced emissions and more effective use of fuel reserves will be substantial. In the longer term, in a hydrogen economy, the SPFC appears to be almost unchallenged as an efficient power producing system.

This paper has outlined the cost and performance targets which must be achieved if the SPFC is to see commercial service. These targets are challenging but probably achievable given time. A successful development programme will demand innovative solutions, worldwide cooperation between organisations with appropriate technical skills, and an emphasis on understanding the science and engineering of these systems.

It must be remembered that fuel cells will have to compete with well-established and reliable products such as the ICE. These are continually being redeveloped and improved based on a full appreciation of their performance and design characteristics. A similar broad base of technical understanding will be required before SPFCs are ready to enter the mass market.

Acknowledgements

Reference

1 D.K. Stevenson, Solid polymer fuel cell systems, Application study to identify and prioritise research and development issues, *ETSU Rep. No. ETSU/FCR/005,* July 1993.