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# A study of SOFC–PEM hybrid systems

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**Abstract**

The benefits of a system combining high- and low-temperature fuel cell types have been assessed using computer predictions. A high-temperature solid oxide fuel cell (SOFC) may be used to produce electricity and carry out fuel reforming simultaneously. The exhaust stream from an SOFC can be processed by shift reactors and supplied to a low-temperature polymer electrolyte membrane (PEM) cell. The overall efficiency predicted for the hybrid system is shown to be significantly better than a Reformer–PEM system or an SOFC-only system. Approximate capital and running cost estimates also show significant benefits compared to the other two systems.
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*Keywords:* Solid oxide fuel cell; Internal reforming; PEM; Hybrid

## 1. Introduction

Because of the interest from transport applications, polymer electrolyte membrane (PEM) fuel cells have made rapid technical progress in recent years. As a result, PEM stack costs are reaching the level at which they are commercially viable for stationary power generation. However, the PEM needs a hydrogen-rich fuel feed, so, in the absence of a cheap and plentiful supply of hydrogen, it is necessary to process other fuels to produce a suitable gas mixture. For this, a reforming operation is required followed by shift reactor operations. The reformer produces a mixture of CO and $H_2$, and, in the presence of $H_2O$, the shift reactors convert the CO to $H_2$. Since the reforming operation is highly endothermic, extra fuel gas must be supplied, thereby reducing the overall system efficiency.

The benefits of combining high-temperature solid oxide fuel cells (SOFCs) with low-temperature (PEM) cells is assessed here. Owing to the internal reforming ability of the SOFC, it is possible to produce both electrical power, and a stream of reformate gas from an SOFC stack. This eliminates the need for a reformer upstream of the PEM, and should result in improved system efficiency. The SOFC exhaust gas is passed through shift reactors to convert most of the carbon monoxide to hydrogen via the shift reaction: $CO + H_2O \rightarrow CO_2 + H_2$. After removal of any residual CO, this hydrogen-rich stream is fed to PEM cells, which produce additional power, thereby increasing the overall efficiency of the system.

The concept of combining fuel cells with other devices is not new. Vollmar and Drenckhahn [1] reported the advantages of placing internal reforming high temperature fuel cells upstream of other processes. Dijkema et al. [2] suggested that fuel cells could be operated on by-product hydrogen produced in the chemical industries. They also advocated 'trigeneration', combining fuel cells with other chemical processes. More recent proposals to use the SOFC for chemical separation of $CO_2$ [3] also recognised that additional benefits may arise from the SOFC rather than simple electricity generation. Finally, much interest is being shown presently in the combination of SOFCs with gas turbines.

## 2. SOFC and PEM stack models

### 2.1. SOFC stack model

In order to simulate hybrid systems combining solid oxide and PEM fuel cells, good stack models for each

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technology are required. A model for an SOFC stack has been developed in which the following assumptions were made:

- The model is able to support the ''Smarter Stack Concept'' (see below)
- Equal mean current densities for the recycle and exhaust anodes
- The reforming reaction is kinetically limited
- The shift reaction is at equilibrium throughout the cells
- Effects transverse to the direction of flow may be ignored
- Plug flow of the gases
- No gas leakage through the electrolyte

The duties of the recycle and exhaust anodes are balanced to give equal current densities. Since ohmic losses in the electrolyte are a source of inefficiency and internal heating, it seems desirable to make the current density as uniform as possible throughout the stack. Under cell conditions the reforming reaction is relatively slow, especially compared to the shift reaction. Hence, it is necessary to include a calculation for the rate of the reforming reaction, while the shift reaction can be assumed to reach equilibrium throughout the stack. Gas flows are assumed to be turbulent so there is substantial transverse mixing. Thus, temperature and composition may be assumed uniform transverse to the direction of flow. Gas leakage through the membrane is neglected because of lack of information. This effect should be small for a flawless electrolyte.

The concept of the Smarter Stack has been described elsewhere [4]. In this concept, two types of anode compartments are used (Fig. 1). Fuel entering the recycle anodes is largely reformed to CO and $H_2$ with only limited electrochemical oxidation. It is then mixed with the fresh fuel gas and recycled. Fuel entering the exhaust compartment is reformed, as well as being electrochemically oxidised to a much greater extent. There are several advantages to this configuration. High Nernst voltages and electrical output are maintained in the recycle cells. Also, the reforming reaction is extended across the cell, hence, creating a more uniform temperature distribution. The model uses a discretised 1D stack method coded in FORTRAN, that is, the stack is divided into a number of increments chosen by the user; transverse variations are ignored. The increments are solved sequentially to produce steady state profiles for the gas composition, temperature, current density and other variables. The following effects are included:

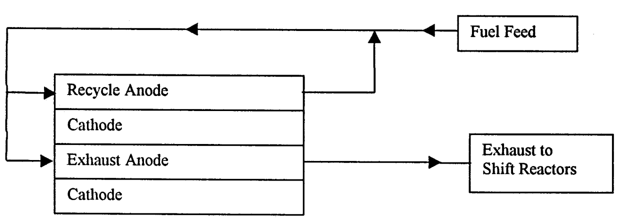


Fig. 1. The ''Smarter Stack'' anode configuration for SOFC.

- Kinetics of the reforming reaction
- Shift equilibrium
- Electrical resistance of electrolyte
- Anode and cathode over-potentials
- Thermodynamic formulae for standard electromotive force and open circuit voltage
- Enthalpy balances at each cell increment to predict the cell temperature profile

Total cell current, cell efficiency and power output are easily denied by summing over all cell increments.

A flag is set to determine which reforming rate expression is used in the SOFC stack model. This feature enables other reforming rate equations to be evaluated when they become available. For this study, all work has been performed with the same reforming rate expression, which has been obtained from work carried out by BG Technology on state-of-the-art SOFC anodes. A flag indicates that the inlet flows are component molar flows in mole per second. The inlet temperatures for the anode and cathode are specified together with the stack outlet temperature and the operating pressure. The total cell area for the recycle and exhaust anodes is given together with the split ratio for the inlet stream. The numbers of cells for the recycle anodes and for the exhaust anodes are specified, followed by the number of increments for the computation.

The model produces a table showing the component molar flows for both types of anodes and for the cathode at six points including the inlet and outlet. The total molar flows and temperatures are also given at the same six points. In addition, the following output information is listed for the recycle and exhaust anodes:

Area ($m^2$)
Volts (V)
Current (A)
Mean current density ($A/m^2$)
Power (W)

Lastly, the following data is listed for the overall cell performance:

Split ratio
Pressure (bar)
Fuel utilisation (%)
Oxygen utilisation (%)
Efficiency (%)

### *2.2. Results from the SOFC stack model*

The model was run at four different areas to determine the effect on fuel utilisation and overall efficiency. The results are shown in Table 1.

Table 1
Results for SOFC model at 1 bar

| Total stack area ($m^2$) | Fuel utilisation (%) | Efficiency (%) | Outlet temperature (°C) |
|---|---|---|---|
| 45 | 0.30 | 20 | 950 |
| 67.5 | 0.40 | 29 | 935 |
| 90 | 0.50 | 36 | 920 |
| 112 | 0.65 | 47 | 910 |

For a fuel utilisation of 0.65%, the results at 3 bar are the same as for 1 bar. However, the cell area is reduced by 5%. At 6 bar and a fuel utilisation of 0.65%, the cell area is reduced by 30% and the outlet temperature increases to 976°C. For a given utilisation, the efficiency does not vary much with pressure.

Information from other workers [5] on SOFC development suggests that the efficiency and fuel utilisation for an SOFC-only system would exceed these predicted values. However, for the hybrid system, the fuel utilisation of the SOFC must be reduced to provide fuel for the PEM. As a result, the SOFC operates at a lower efficiency in the hybrid system. However, less cell area is required, so there is a capital cost benefit compared to the SOFC-only system. At present, the sensitivity of the predictions with respect to the reforming rate correlation is not known, but this effect needs to be investigated further when the appropriate information becomes available.

### 2.3. PEM fuel cell stack model

A PEM fuel cell model was constructed along similar lines to the SOFC stack model. The following assumptions were made to develop the model:

- Isothermal operation, that is, no heat balance calculations are performed
- Constant vapour pressure of water throughout the cell
- Effects which are transverse to the direction of flow may be ignored
- Plug flow of the gases
- No gas leakage and water transport through the membrane

Since a PEM cell is cooled to remove waste heat, it is not unreasonable to assume isothermal operation. This simplifies calculations by making heat balance equations unnecessary. The vapour pressure of water is mainly a function of temperature so a constant value is assumed in accordance with the assumption of isothermal operation. Gas flows are assumed to be turbulent so there is substantial transverse mixing. Thus, temperature and composition may be assumed uniform in any direction transverse to the flow. Gas leakage and water transport through the membrane are neglected because of lack of information. These effects should be small for a sound membrane.

The PEM model is also a discretised 1-d model that has been coded in FORTRAN, that is, the stack is divided into a number of increments chosen by the user and transverse variations are ignored. The increments are solved sequentially to produce steady state profiles for the gas composition and current density for a given overall operating voltage. For each increment, the Tafel equation and its derivative are used to determine the current density that matches the specified operating voltage, that is

$$E = E_0 - b\log i - Ri$$

or

$$\frac{\partial E}{\partial i} = -\frac{b}{i} - R$$

where $E$ = cell potential, $E_0$ = cell electromotive force, $i$ = current density, $b$ = constant in voltage units, $R$ = constant in resistance units.

In order to solve this equation, the model uses correlations to calculate the following data at each increment:

- Electrical resistance of polymer
- Cathode over-potentials
- Standard electromotive force
- Open circuit voltage
- Water flow

Thus, $E$, $b$ and $R$ can be derived. For each increment, the cell potential $E$ is determined and compared with the desired voltage operating point. The derivative form is then used to vary the current density to match the desired voltage operating point. The vapour phase water content is found from the vapour pressure and the total pressure. The water flow is then calculated using the flows of the other gases. The flow of water in liquid form is then deduced by subtraction. Total cell current, cell efficiency, and power output are easily obtained by summing up all cell increments.

Input streams may be specified as component flows in mole per second or as a molar composition followed by an overall molar flow. For the anode and the cathode, the input streams consist of the molar flows of $CH_4$, CO, $CO_2$, $H_2$, $H_2O$, $O_2$, and $N_2$. The operating temperature, total pressure, and water vapour pressure are needed, as well as the cell operating voltage and cell active area.

Table 2
Comparison of models at 1.4 bar and 90 $m^2$ geometric area

| | Model | |
|---|---|---|
| | Empirical model | Discretised PEM model |
| Operating pressure (bar) | 1.4 | 1.4 |
| Total active area ($m^2$) | 90 | 90 |
| Fuel utilisation (%) | 0.714 | 0.517 |
| Operating voltage | 0.7657 | 0.7657 |
| Current density ($A/m^2$) | 2185 | 1584 |
| Power (kW) | 150.6 | 109.2 |

As for the SOFC model, the PEM model tabulates the results from the stack. The component gas molar flows for the anode and cathode are given at six points in the stack including the inlet and outlet. The table also includes the total molar flow and water flow at each of the six points. After the table, the following information is listed:

Cell area ($m^2$)
Operating voltage (V)
Current (A)
Mean current density ($A/m^2$)
Power (W)
Pressure (bar)
Fuel utilisation (%)
Oxygen utilisation (%)
Efficiency (%)

### 2.4. Results from the PEM model

For the purpose of the systems analysis described in Section 3, an empirical PEM stack model was also used. Full details of this model are not available, but it is based on correlations of stack performance measurements obtained from published data. It does not use discretisation to track compositional changes through the stack. However, the opportunity has been taken to compare the results from the two models for a range of pressures and cell areas. For this comparison, three overall pressures and two cell areas have been chosen. Some results are shown in Tables 2–4.

In all cases, the discretised PEM model underestimates the performance predicted by the empirical model. The operating voltages are identical because they are required to define the load; they are part of the input data. Good agreement between the model and data was not expected because no validation of the coefficients of the Tafel equation had been carried out.

In its present form, the model is an implementation of an algorithm rather than a validated tool. As such, the model is satisfactory, but steps must be taken to calibrate it against experimental data, as these become available.

The algorithm used in the empirical model computes cell voltage for a given cell area and fuel utilisation. The PEM model computes fuel utilisation and efficiency from cell voltage and cell area. In other words, the discretised PEM model will give a larger fuel utilisation if the cell area is increased, but the empirical model will not. This is a limitation of the empirical model because it assumes that a given performance can be achieved for a given cell area.

It is desirable to include a heat balance calculation in order to predict the cell temperature profile and cooling duty. At the same time, a correlation for water partial pressure, in terms of temperature, should be included. However, both of these enhancements are secondary to improving the calibration of the electrochemical calculations.

Table 3
Comparison of models at 1.4 bar and 72 $m^2$ geometric area

| | Model | |
|---|---|---|
| | Empirical model | Discretised PEM model |
| Operating pressure (bar) | 1.4 | 1.4 |
| Total active area ($m^2$) | 72 | 72 |
| Fuel utilisation (%) | 0.714 | 0.584 |
| Operating voltage | 0.7141 | 0.7141 |
| Current density ($A/m^2$) | 3642 | 2236 |
| Power (kW) | 140.4 | 115.0 |

Table 4
Comparison of models at 3.0 bar and 90 $m^2$ geometric area

| | Model | |
|---|---|---|
| | Empirical model | Discretised PEM model |
| Operating pressure (bar) | 3.0 | 3.0 |
| Total active area ($m^2$) | 90 | 90 |
| Fuel utilisation (%) | 0.714 | 0.422 |
| Operating voltage | 0.7895 | 0.7895 |
| Current density ($A/m^2$) | 2185 | 1289 |
| Power (kW) | 155.2 | 91.6 |

## 3. Systems study

With the present information, the system configuration shown below is thought to be the best (Fig. 2). Natural gas enters the SOFC section where reforming and electrochemical oxidation occur. The SOFC stack produces electrical power together with an exhaust stream containing unused CO and $H_2$. This exhaust stream is cooled and passed to the shift reactors in which the CO reacts with $H_2O$ to produce $CO_2$ and $H_2$. There is sufficient $H_2O$ in the stream to convert all the CO, provided the SOFC fuel utilisation exceeds 0.5%. When operating at utilisations below this level, water would need to be injected and recovered downstream. In this way, only a small make-up water supply would be needed. After the shift reactors, the remaining traces of CO are removed by selective catalytic oxidation. This is necessary to prevent poisoning of the catalysts used in the PEM stack. The resulting $H_2$-rich stream is cooled to about 70°C before entry to the PEM

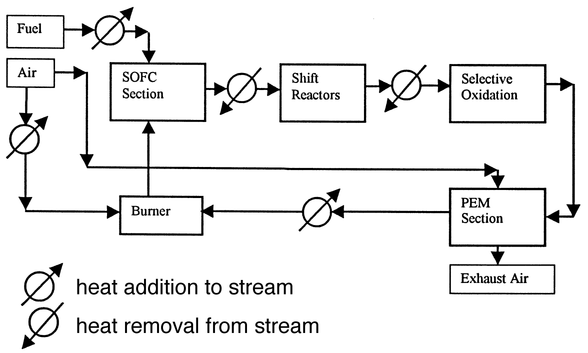


Fig. 2. Schematic of system configuration.

section. As the anode stream from the PEM section contains unused $H_2$, it is reheated and combusted using the air stream to the SOFC cathode. This utilises the unused fuel energy.

In order to optimise the process, several factors must be investigated. Clearly the fuel utilisation must be as high as possible, but its distribution between the SOFC and the PEM is also important. If other variables are unchanged, a high fuel utilisation can only be obtained by increasing cell area, thereby increasing capital cost. For a given area of cell, SOFCs are more expensive than PEMs, but they are usually more efficient, so the optimum distribution of fuel utilisation is not a straightforward matter. Furthermore, if no steam is supplied to the SOFC, the minimum fuel utilisation is 25% when using $CH_4$, otherwise insufficient $H_2O$ is produced to reform all the incoming hydrocarbon. Moreover, to avoid the need to add extra steam before the shift reactors, this fuel utilisation must be increased to 50%. Another important issue in process optimisation is the design of the heat recovery processes. There is a large cooling duty associated with the streams leaving the SOFC and the shift reactors. For good system thermal efficiency, this heat must be transferred effectively to colder streams such as:

- The incoming air and fuel
- The anode exhaust from the PEM

The process has been simulated using the HYSIS flowsheet package and the empirical PEM model which is interfaced to HYSIS. This simple model has been chosen instead of the discretised model described above because it has been validated against cell performance measurements. A simplified model for the SOFC, which could be implemented with standard HYSIS unit operations, to enable a complete simulation of the process, has been used. This SOFC model makes the following assumptions:

- All the hydrocarbon fuel is reformed because the outlet temperature is sufficiently high.
- The shift reaction is at equilibrium because the kinetics are fast.
- The oxygen requirement is defined by the pre-determined fuel utilisation.

Subsequently, detailed modeling of the SOFC has been carried out, using the model described in Section 2, to predict its power output and determine the overall system power and efficiency.

Process studies have been carried out at the following pressure levels in order to determine the effect of pressure on system performance:

- Low pressure (2 bar falling to 1 bar)
- Medium pressure (3.6 bar falling to 2.88 bar)
- Higher pressure (6.6 bar falling to 5.88 bar)

Table 5
Summary of power figures

| | |
|---|---|
| SOFC power (kW) | 369.3 |
| PEM power (kW) | 146.7 |
| Turbine power (kW) | 100.3 |
| Compressor power (kW) | −100.8 |
| Net power output (kW) | 515.5 |
| Electrical output (kW) | 489.7 |
| Overall efficiency (%) | 61 |

These pressure levels represent the limits for PEM operation with present knowledge. A turbine is used to recover power from the SOFC cathode exhaust stream. This is of particular importance when operating at increased pressure. Full details of the system configuration and simulation are given in a report presently being produced for the DTI. During the process study, the effects of altering the utilisations for the SOFC and PEM cell have also been investigated.

Results from the systems study showed that best performance is obtained for the medium pressure system. For this case, ignoring the SOFC, the net system power output for a PEM active area of 54 $m^2$ is 146.7 kW. From the SOFC computer model, a stack efficiency of 46% is predicted for the medium pressure conditions. The fuel inlet flow is 1 mole/s of methane, which provides 802.7 kW of chemical energy, that is, the SOFC power output is 369.3 kW. Allowing an inverter efficiency of 0.95%, the power figures are summarised in Table 5.

The low-pressure system gives a similar net power so it does not appear essential to pressurise the system. However, for the low-pressure system, the SOFC air must be compressed to 2 bar and the PEM air to 1.5 bar. Two gas compressors are included for these operations. If the pressure losses in the system could be reduced sufficiently it would be possible to use blowers instead of compressors resulting in a capital cost reduction. For the medium pressure system, the corresponding air pressures are 3.65 and 3.05 bar. To achieve these pressures would require more expensive equipment than the low-pressure system.

## 4. Cost analysis

In fuel cell technology, the closest competitors for the hybrid system are the Reformer–PEM system and the SOFC-only system. The major differences between these systems are in the costs of the stacks and reformer, and the cost of fuel, so, for present purposes, the heat exchanger costs were ignored. Assuming that the cost of the other balance of plant items is roughly the same in all three cases, Table 6 gives very approximate estimates of these costs for a 200-kWe system. All these costs are based on future projections found in the literature and should not be quoted out of context. The figures have been calculated for

Table 6
Comparative system costs

| System type | SOFC–PEM | Reformer–PEM | SOFC[a] |
|---|---|---|---|
| SOFC stack ($) | 73 000[b] (26 000) | | 142 000 (51 000) |
| Reformer ($) | | 35 000[c] (22 000) | |
| PEM stacks ($) | 11 000 (7000) | 21 000[d] (13 000) | |
| Fuel cost ($)[e] | 561 000[f] | 855 000[g] | 653 000[h] |
| Total ($) | 645 000 | 911 000 | 795 000 |

[a] The stack cost and system efficiency are based on simulations using the same SOFC stack model used in the SOFC–PEM system simulation.
[b] This assumes a stack life of 5 years and a cost of US$550/$m^2$ [5]. Note that the SOFC stack costs in this system are much less than in a normal system because the stack is small owing to the higher current density and the lower current.
[c] This figure is based on the BG compact reformer target cost of £67/kW, and a life of 10 years (£1 = US$1.64).
[d] This assumes the Arthur D. Little figure of US$65/kW, and a life of 10 years.
[e] A natural gas cost of US$0.02/kW h is assumed, with an annual reduction of 2%.
[f] Assuming a system efficiency of 64% calculated above.
[g] Assuming a system efficiency of 42%.
[h] Assuming a system efficiency of 55%.

a 20-year system life, and are net present values, that is, the equipment replacement costs are included. A cash flow discount rate of 5% has been assumed which is not especially optimistic. Figures in brackets are initial capital costs.

Table 6 shows that the SOFC stack will cost more than a reformer, but the difference is easily offset by the reduced fuel consumption. The heat exchanger costs, which have not been included, will be the lowest for the Reformer–PEM system.

## 5. Conclusions

From the computer simulations of the combined SOFC–PEM system, the following performance figures have been obtained:

(a) Overall system efficiency: 61%
(b) Net electrical output: 489.7 kWe

By comparison, the overall efficiency of a Reformer–PEM system is in the range 37–42%, based on computer predictions.

Rough capital and running cost estimates show that the hybrid system has a significant financial benefit compared to the SOFC-only system, and a substantial benefit compared to the Reformer–PEM system. However, it should be emphasised that these are only very rough initial estimates and should be reviewed as further information on stack costs becomes available.

Among others, the following factors are significant for the optimisation of the hybrid system:

(a) The distribution of the power output between the SOFC and PEM stacks
(b) The effectiveness of the heat recovery processes within the system

The medium pressure system gives the highest overall efficiency, but it is, at best, less than 5% better than the low-pressure system. The high-pressure system gives the lowest efficiency. Thus, the benefits of operating at increased pressure are not proven. However, it does seem easier to maintain humidification of the PEM at higher pressures.

Using methane as the fuel, without added steam, the lower limit on the SOFC fuel utilisation is 25%. At lower levels, insufficient $H_2O$ is produced to fully reform the methane in the SOFC. Regarding the design of the shift reactors, there are benefits in increasing the SOFC utilisation or injecting steam upstream of these reactors. The system can be designed to be self-sufficient in water.

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