

## HIGH-TEMPERATURE SOLID OXIDE FUEL CELL — TECHNICAL STATUS

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### Summary

The high-temperature Solid Oxide electrolyte Fuel Cell (SOFC) represents the basic building block for power generation in a variety of applications, ranging from total energy systems for residences, industrial cogeneration systems, and utility central station power production. The cell operates at approx. 1000 °C, using a variety of fuels. Yttria-stabilised zirconia is the solid electrolyte that conducts oxygen ions from the cathode to the fuel electrode where the fuel is oxidised to release electrons (current) to an external load.

This paper describes the operating principle of the SOFC and relates its component composition in the thin layer concept. Performance and life test data to 5000 h will be presented. Sulphur tolerance prediction data and actual tolerance test data are also presented. In addition, test results will include the effect of various fuels, *e.g.*, CO and/or H<sub>2</sub> (as derived from coal gas), on the performance of the SOFC cell. The status of the present technology is also described. Finally, the usefulness of the SOFC generator, studied as part of a cogeneration system is discussed.

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### Historical background

About 1900 Nernst conceived the idea that oxidic conductors could be used as light sources in lamps, in place of carbon filaments. He discovered that the low conductivity of pure zirconium oxide could be improved by adding other oxides. He found that  $(Y_2O_3)_{0.15} (ZrO_2)_{0.85}$  was the most promising composition in this respect [1]. Wagner then determined that vacant oxygen lattice sites impart mobility to oxygen ions in this imperfect fluorite structure and found that the substitution of zirconium ions by others of lower valence, *e.g.*, yttrium or calcium, proportionally increase vacant oxygen ion lattice sites [2].

Ruka and Weissbart in 1958 conceived the idea that stabilised zirconia, with good oxygen ion conductivity at elevated temperature, could serve as a solid electrolyte in a high-temperature solid oxide fuel cell. They reconfirmed that oxygen ion conductivity existed in calcia-stabilised zirconia and

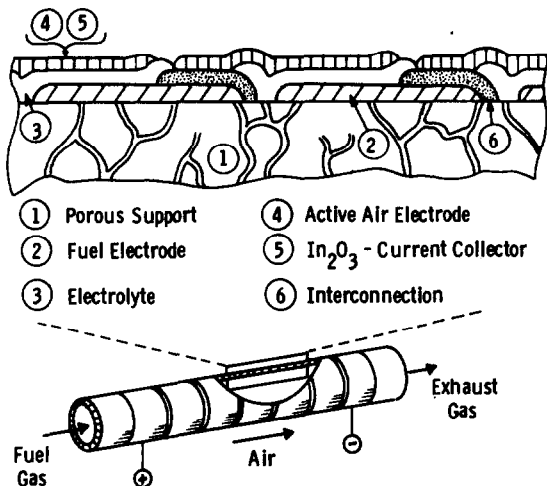


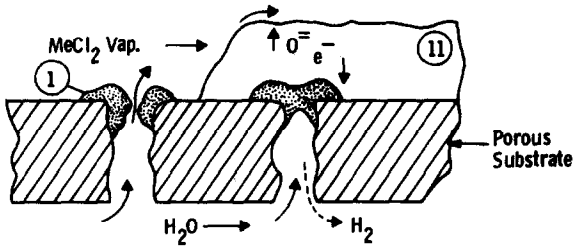
Fig. 1. Schematic diagram of the solid oxide series connected fuel cell stack.

measured its conductivity as a function of temperature up to 1000 °C. These authors constructed vacuum tight cells having solid oxide electrolyte  $(\text{ZrO}_2)_{0.85} (\text{CaO})_{0.15}$  and platinum electrodes, established  $V-I$  curves with various fuels and confirmed the theoretical voltage relationship of the cell [3].

Serious development efforts began on the high temperature Solid Oxide Fuel Cell (SOFC) in 1962 and continued without interruption to 1970. The objective of this work was to develop a commercial fuel cell power generating system which would use coal as a fuel source. The thin film fuel cell concept (Fig. 1), in which a porous support tube of calcia-stabilised zirconia (in the cubic phase) was used as a structural member, was developed.

The fuel electrode, the electrolyte, the air electrode, the porous support tube composition, and useful fabricating processes were identified. The electronically-conducting interconnection material was still a problem and, in 1970, presented the most singular major obstacle to construction of a working SOFC fuel cell. Other problem areas included the development of fabrication techniques for producing gas tight thin films of electrolyte and interconnection, especially in their overlap regions, to insure both separation between the fuel and oxygen at 1000 °C (*i.e.*, to attain near theoretical open circuit potential and to avoid adverse reactions with component materials) and good cell performance. Because the OCR/DOI contract was for the construction and demonstration of an SOFC generator and did not result in a practical cell that could demonstrate performance and life, the program was terminated in 1970.

In 1970 Isenberg, in an independent investigation, conceived and developed the electrochemical vapour deposition (EVD) process (Fig. 2). This uniquely insures the fabrication of gas tight layers of electrolyte and interconnection materials, as well as gas tight sealing at their overlap region.



Phase I — Pore Closure by CVD  
 $\text{MeCl}_2 + \text{H}_2\text{O} \rightarrow \text{MeO} + 2 \text{HCl}$

Phase II — Scale Growth by EVD  
 $\text{MeCl}_2 + \text{O}^= \rightarrow \text{MeO} + \text{Cl}_2 + 2\text{e}^-$   
 $\text{H}_2\text{O} + 2\text{e}^- \rightarrow \text{H}_2 + \text{O}^=$

Fig. 2. Principle of electrochemical vapour deposition, EVD.

In 1978, as a part of the U.S. National Fuel Cell Programme, a 2-year programme resulted in the identification of a suitable interconnection material and successful fabrication and testing of a 3-cell stack that operated for 700 h, at  $200 \text{ mA cm}^{-2}$  at  $1000^\circ \text{C}$  with 80% voltage efficiency, using hydrogen fuel and air as the oxidant source. The results of a further 2-year programme and the status of the present programme are discussed here.

### The cell and its operating principle

Table 1 presents the 1978 vintage and present SOFC component design and related fabrication processes. The advantage of the present design is that metallic contacts, which are protected by the reducing environment of the fuel stream, are made to the metallic component (Ni or Co) of the fuel electrode. This design also leads to a feasible generator design concept in which metallic leads can be used in series and/or parallel connections of fuel cell bundles. The key to successful operation of the cell is that all components must be compatible and ensure stable interfaces at  $1000^\circ \text{C}$  for prolonged operating times. All components must have thermal expansions ( $10.5 \times 10^{-6} \text{ }^\circ \text{C}^{-1}$ , RT -  $1000^\circ \text{C}$ ) closely matched to that of the porous support tube, to minimize or eliminate stresses due to differential thermal expansion between components. Further, no fabrication processes must adversely affect those components already assembled onto the porous support tube as the cell is being constructed. All components listed in Table 1 meet these requirements.

The voltage of the SOFC is dependent on the difference between the oxygen partial pressure in the fuel and in the air streams. This is, generally, about 17 orders of magnitude and is given by the Nernst equation:

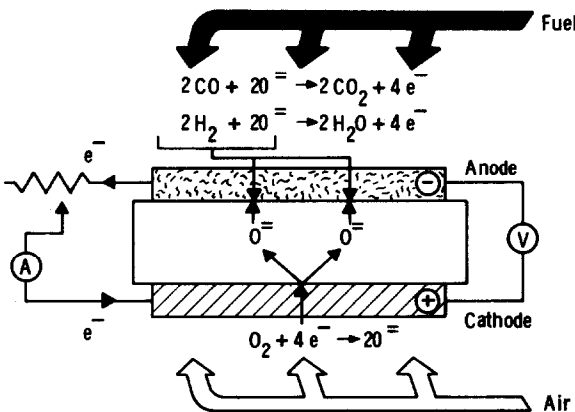
**TABLE 1**  
Solid oxide fuel cell — summary of components, materials and fabrication processes

Component	Material	Fabrication process
<i>Series-cell stack</i>		
Support tube	calcia-stabilised zirconia	extrusion/sintering
Fuel electrode	nickel-zirconia cermet cobalt-zirconia cermet	slurry coat/sinter
Solid electrolyte	yttria-stabilised zirconia	electrochemical vapour deposition (EVD)
Interconnection	modified lanthanum chromite	EVD
Air electrode	tin-doped indium oxide current collector over porous zirconia	CVD In <sub>2</sub> O <sub>3</sub> and impregnation
<i>New cell design</i>		
Support tube	calcia-stabilised zirconia	extrusion-sintering
Air electrode	modified lanthanum manganite	slurry coat/sinter
Solid electrolyte	yttria-stabilised zirconia	EVD
Interconnection	modified lanthanum chromite	EVD
Fuel electrode	nickel-zirconia cermet cobalt-zirconia cermet	slurry coat/sinter

$$E = \frac{RT}{nF} \ln \frac{pO_2 \text{ (air stream)}}{pO_2 \text{ (fuel stream)}} \tag{1}$$

In effect the SOFC is an oxygen concentration cell. At 10<sup>-17</sup> atmosphere of oxygen in the fuel stream (and 0.21 atmosphere oxygen in the air side) the theoretical open circuit voltage at 1000 °C is 1.03 V.

The operating principle of the SOFC is presented in Fig. 3. When an external load is applied to the cell, oxygen from the air is reduced at specific reaction sites in the porous air electrode to produce oxygen ions. These ions then readily migrate at 1000 °C through the solid electrolyte to the fuel electrode. At specific sites on the porous fuel electrode the fuel, H<sub>2</sub> (or CO + H<sub>2</sub>),



**Fig. 3.** Schematic diagram of the solid oxide fuel cell, indicating how oxidation of the fuel generates electric current to the external load.

for example, is oxidized to produce  $\text{H}_2\text{O}$  (or  $\text{CO}_2 + \text{H}_2\text{O}$ ). The number of oxygen ions reacting at the fuel electrode equals the number of oxygen ions entering the electrolyte, so that the electrical neutrality of the electrolyte is maintained. Electrons, released by this oxidation, flow through the external load. This reaction continues as long as fuel and air are supplied. Up to  $\sim 90\%$  fuel utilisation can be expected in operating this cell at an average cell current density of  $400 \text{ mA cm}^{-2}$ . Some residual fuel must remain unburnt in the exhaust stream from the fuel cell to protect metallic components both in the fuel electrode and in the electrical connectors. However, this residual fuel can be used in a heat exchange to preheat incoming air and/or fuel. The exhaust gas from the fuel cell is at  $900 - 1100^\circ\text{C}$  and can be used for producing process steam or in a steam turbine bottoming unit to produce more electricity.

### Technology status

A major breakthrough in proving the SOFC cell technology occurred when a 7-cell stack (series connected) demonstrated  $\sim 5000$  h of life at  $1000^\circ\text{C}$ , under a variety of operating conditions. Figure 4 presents the test results of this cell. For approx. 3000 h the stack operated at  $1000^\circ\text{C}$  with  $\text{H}_2$  fuel and generated  $400 \text{ mA cm}^{-2}$  of current at  $\sim 0.72 \text{ V/cell}$ . During the period it was subjected to 11 thermal cycles ( $1000^\circ\text{C}$  - room temperature -  $1000^\circ\text{C}$  in 4 h). The stack was then tested in a simulated coal gas, "spent fuel" condition ( $15\% (2\text{CO}:1\text{H}_2) - 85\% \text{CO}_2$ ) at  $100 \text{ mA cm}^{-2}$ : it operated steadily at  $\sim 0.67 \text{ V/cell}$  for approx. 400 h. For the next 700 h the stack operated at  $150 \text{ mA cm}^{-2}$  under these same fuel conditions at  $\sim 0.62 \text{ V/cell}$ . Then 50 ppm  $\text{H}_2\text{S}$  impurity was introduced to this fuel composition and the stack was operated for an additional 800 h. The only effect caused by the sulphur impurity was an immediate loss of about 5% in operating cell voltage, after which the stack voltage remained constant. On removal of the sulphur impurity from the fuel stream, the stack voltage finally returned to  $\sim 0.62 \text{ V/cell}$  (Fig. 4).

Previous work had predicted, prior to this test, that up to 90 ppm and 200 ppm  $\text{H}_2\text{S}$  could, theoretically, be safely tolerated by nickel and cobalt, respectively, at  $1000^\circ\text{C}$  and at  $\sim 0.70$  open circuit cell voltage (Fig. 5). Post-test examinations of the cell components and their interfaces from this seven-cell stack indicated no interdiffusion effects, as might be caused by cation migration, had occurred and that the structures were unchanged. Because of this test, confidence has been gained in expecting long life for the SOFC. Great difficulties were encountered in trying to design generators with the cell structure described so far. The main problem was to establish electrical contact between cell stacks on the air side. This required either noble metals or bulky oxide conductors. Without abandoning processing methods and the thin layer approach, a new cell configuration was used where the air electrode is next to the support tube. This allows contact to

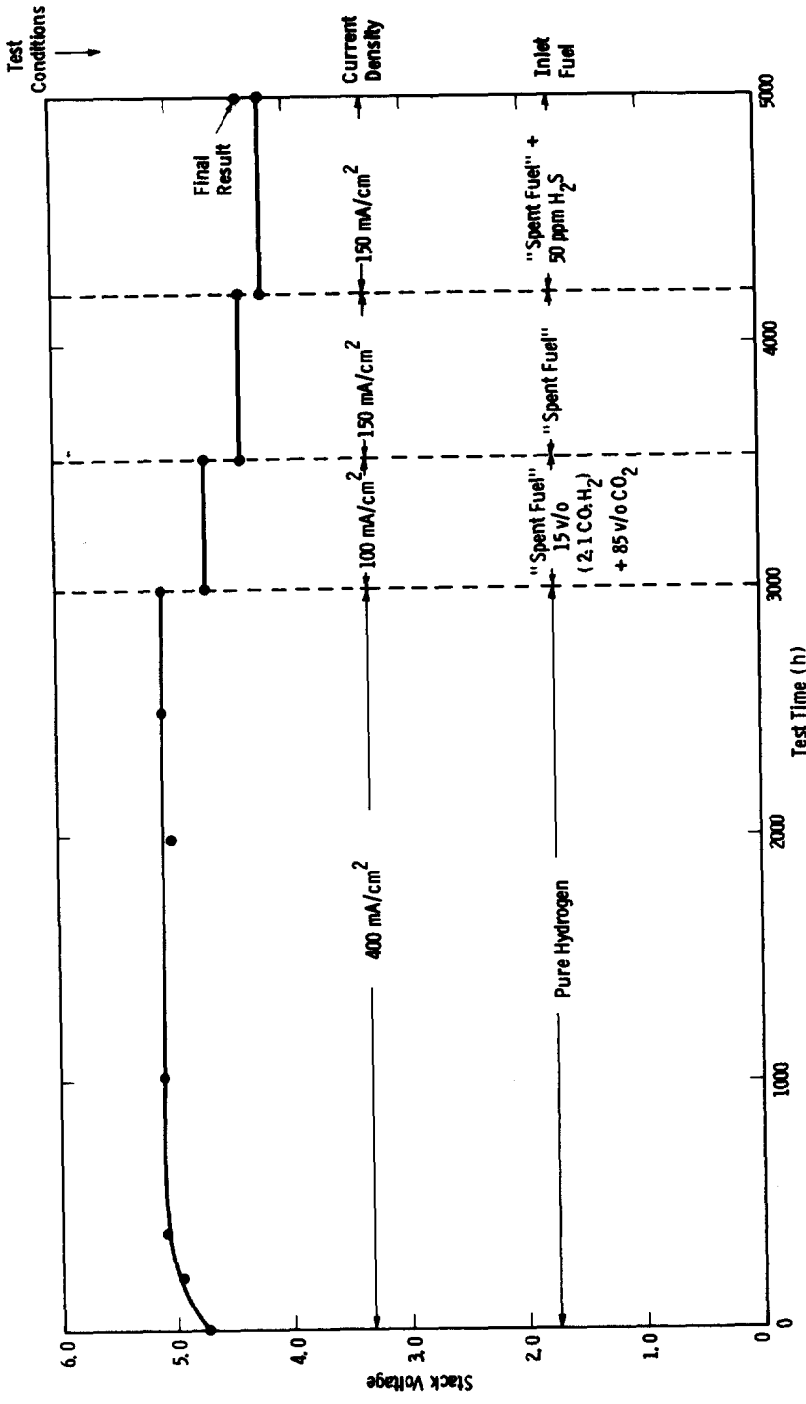


Fig. 4. Life performance at 1000 °C of a series-connected 7-cell SOFC stack.

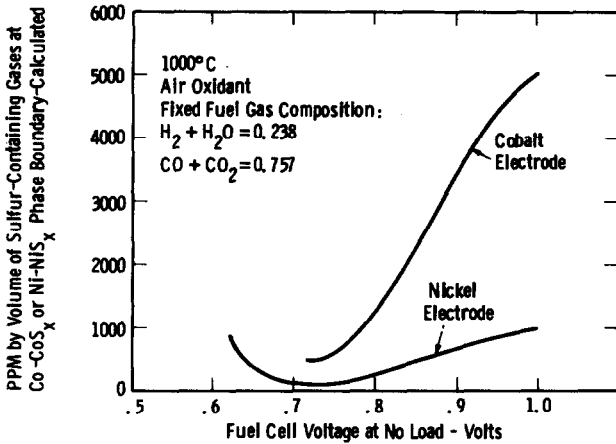


Fig. 5. Concentration of sulphur-containing gases at which nickel and cobalt cermet fuel electrodes will begin to sulphide as a function of the fuel cell voltage.

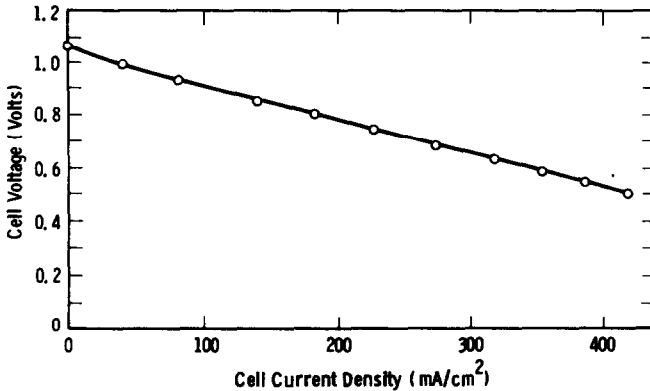


Fig. 6. Performance of the new design cell at 1000 °C,  $H_2$  fuel, air as the oxidant source.

both the fuel electrode (directly) and the air electrode (*via* an interconnection) in the reducing fuel atmosphere with low cost metallic conductors. This has required changes in processing sequences: the new materials for the new cell design are listed in Table 1.

Figure 6 presents performance data obtained on the new cell design. It has delivered  $0.20 \text{ W cm}^{-2}$  at  $400 \text{ mA cm}^{-2}$  with excess fuel and, when fully developed, the performance of this new cell is expected to match that obtained in cells of the series-stack design, described earlier.

### System conditions

The design, cost, and benefit of an industrial cogeneration system, using an SOFC generator, has been studied by Federmann *et al.* [6]. An integrated

aluminium production plant was selected for the study, which was based on the electrical and thermal demand of that plant and its correlation to the electrical and thermal output of a 220 MW<sub>e</sub> SOFC generator. The cogeneration system was designed to the degree necessary to provide a preliminary analysis of economic and technical viability, when compared with the use of conventional energy sources in the year 1990. The system was not optimised nor was any change made in the aluminium production process. Three variations in cogeneration were considered and were based on the use made of the fuel cell exhaust heat (assumed at 800 °C) from a 220 MW<sub>e</sub> DC SOFC generator. In the first mode, only process steam was produced. In the second mode, exhaust heat was used to produce some steam and some electrical power. In the third mode only a.c. electric power was produced. These systems are shown in Figs. 7 - 9 [7].

Table 2 lists the assumptions used in the study under assumed "base case" conditions for the 3 systems. These included: electric and gas\* prices, projected escalation at the time of installation (1990), SOFC system capital cost and life. Also shown in Table 2 are the general economic factors which were included in the study; inflation rate, discount rate, and investment tax credit. A cash flow analysis was made of the SOFC systems and compared with cash flow for projected fuel purchase. A "viability ratio" (V.R.) was developed for this study [6], where

$$V.R. = \frac{\text{Present Value Fuel Cell System Cash Flow}}{\text{Present Value Fuel Purchase Cash Flow}}$$

over a 20 year period. A V.R. of less than 1 indicates a saving and the degree of saving is 1 minus the V.R. value.

Table 3 presents the effect of varying some of the assumed economic values of the base case which gave a viability ratio of 0.7. It can be seen that changes in discount rate, projected inflation rate and investment tax credit make only minor changes in the V.R. Table 4 indicates the sensitivity of the electric and gas rates for the 3 cogeneration systems studied and the base case is highlighted. Figures 10 and 11 [6] summarize, in "sensitivity carpet plots", the effect of electric, gas, and SOFC fuel cell generator cost on the viability ratio with 2% and 0% electric cost escalation. The centre dot is the base case. These plots indicate that cost of electricity is the most sensitive factor in determining the V.R. The effect of variation in the initial cost of the SOFC generator is minimal, even at ±50% of that cost. The SOFC generation system could, therefore, significantly offset the escalating cost of producing aluminium in an integrated plant.

## Conclusions

Solid oxide fuel cell technology has advanced to the stage where component materials and processes have been defined. Fabricated cells have

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\*Derived from coal gasification.



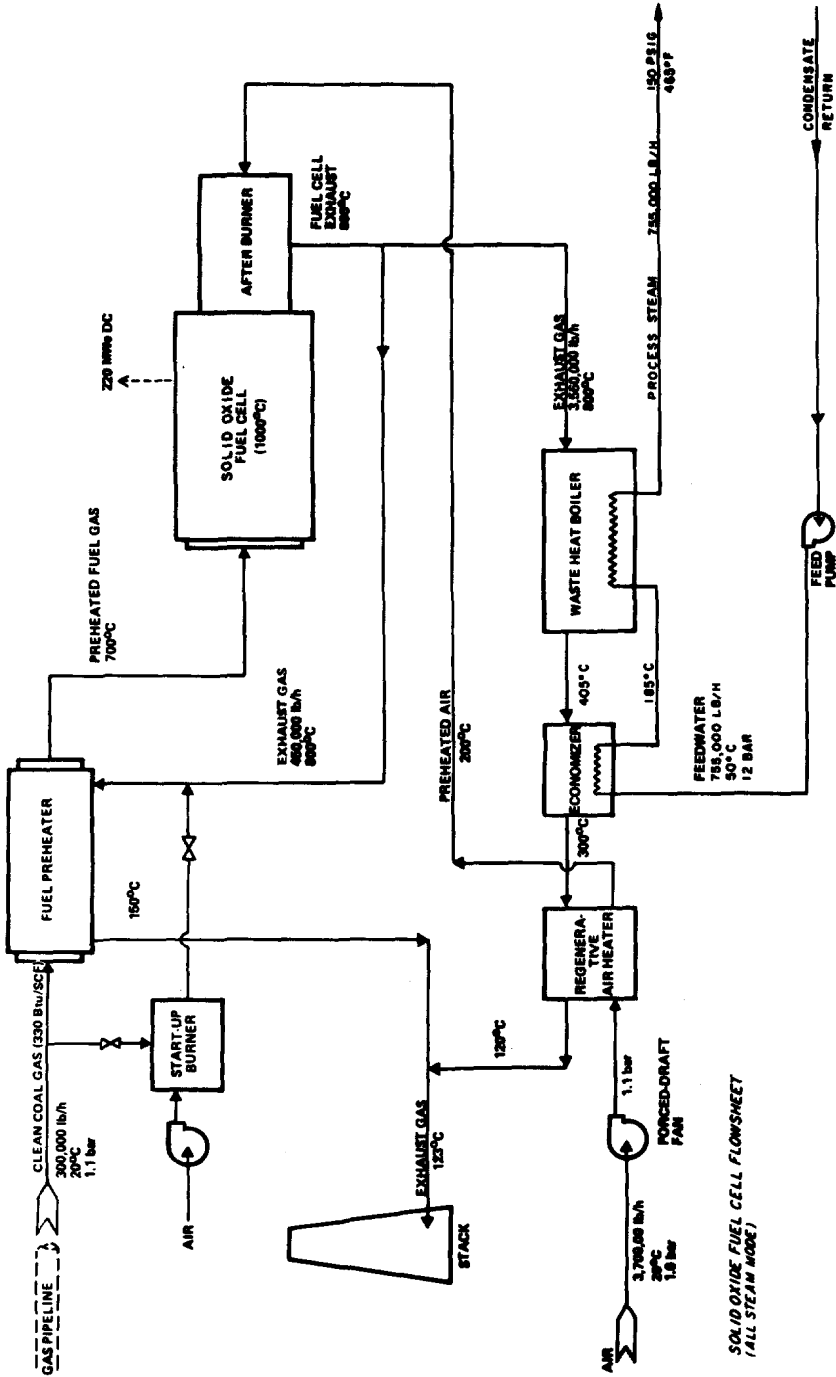


Fig. 7. D.c. power and all process steam using 220 MW<sub>e</sub> d.c. SOFC generator.

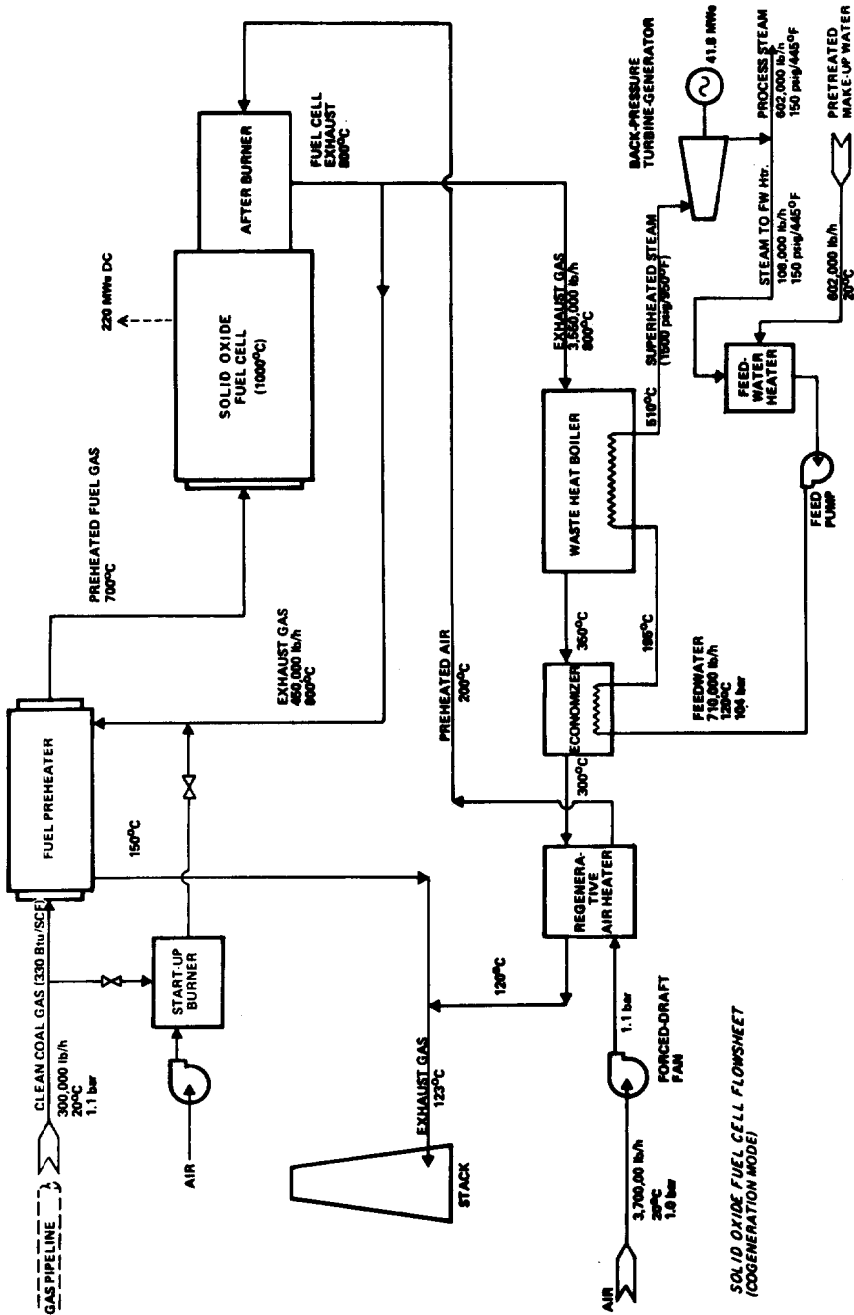


Fig. 8. D. c. power and process steam/a.c. power using a 220 MW<sub>e</sub> d. c. SOFC generator.

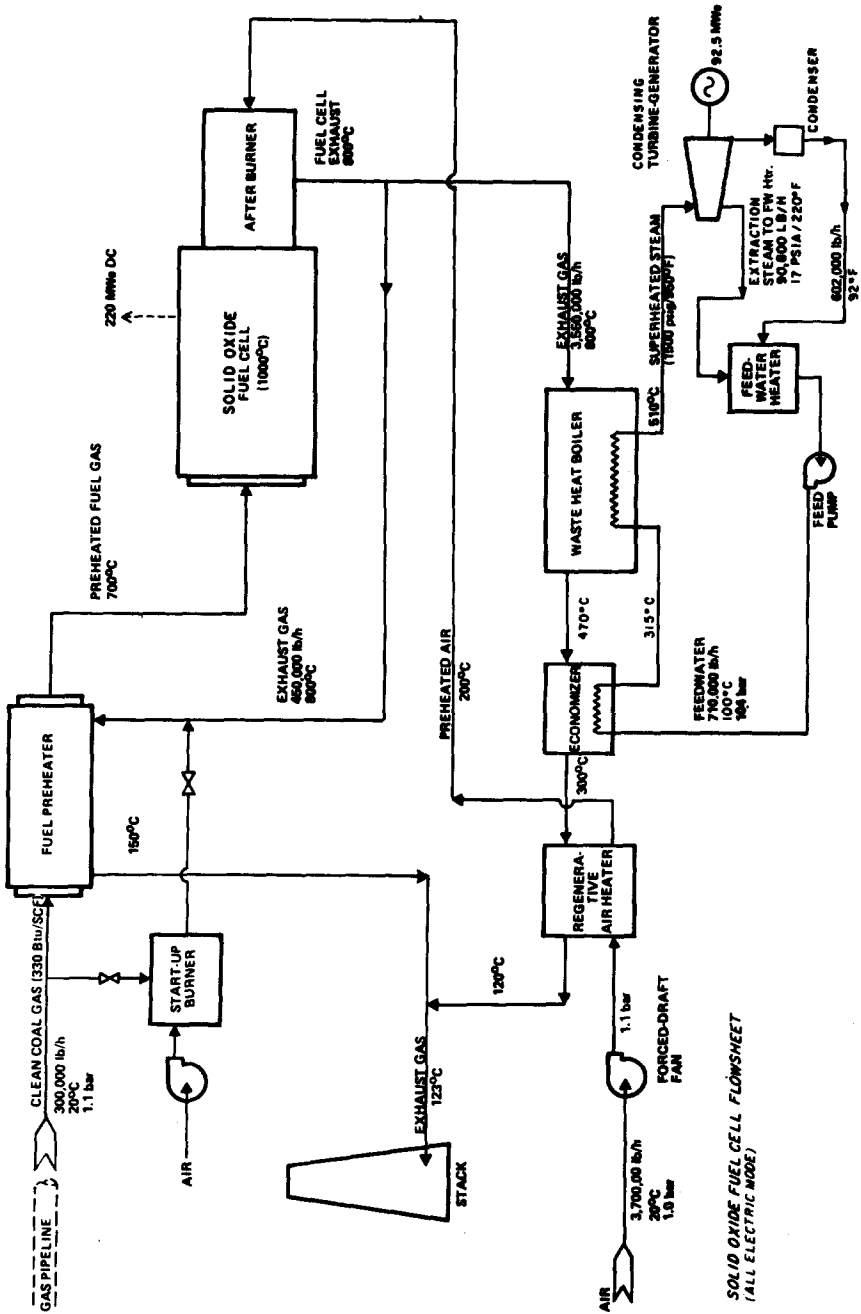


Fig. 9. All electric power using a 220 MW<sub>e</sub> d.c. SOFC generator.

TABLE 2

Base case assumptions (1990 costs in 1980 dollars)

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\$0.05/kW h electric, 2% escalation over inflation  
 \$6/10<sup>6</sup> BTU gas, 0% escalation over inflation, thermal value: \$6/10<sup>6</sup> BTU

Capital cost:

(1) SOFC fuel cell generator + thermal (220 MW <sub>e</sub> , 885 × 10 <sup>6</sup> BTU/h thermal)	= \$120 × 10 <sup>6</sup>
(2) SOFC fuel cell generator + a.c. electric + thermal (261 MW <sub>e</sub> , 731 × 10 <sup>6</sup> BTU/h thermal)	= \$146 × 10 <sup>6</sup>
(3) SOFC fuel cell generator + all electric (312 MW <sub>e</sub> )	= \$166 × 10 <sup>6</sup>

5 Y life = 20 Y plant operation  
 6% inflation, 10% discount rate  
 15% investment tax credit

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TABLE 3

Economic variations. Base case = 0.70 viability ratio

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SOFC replacement every five years	= 0.69
No investment tax credit	= 0.73
15% discount rate	= 0.73
10% inflation, 15% discount rate	= 0.72
\$4/10 <sup>6</sup> BTU thermal value	= 0.76

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TABLE 4

Comparison of three systems

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Electric			Gas			
\$/kW h	Esc. (%)	Esc. (%)	\$4/10 <sup>6</sup> 0	BTU 2	\$6/10 <sup>6</sup> 0	BTU 2
0.03	0	S	1.06	1.21	1.24	1.40
		C	1.02	1.20	1.22	1.42
		E	1.19	1.55	1.62	2.16
0.03	2	S	0.83	0.98	1.00	1.16
		C	0.78	0.94	0.96	1.15
		E	0.84	1.09	1.13	1.51
0.05	0	S	0.74	0.88	0.90	1.07
		C	0.69	0.83	0.86	1.04
		E	0.72	0.93	0.97	1.29
0.05	2	S	0.56	0.68	0.70	0.85
		C	0.51	0.63	0.65	0.80
		E	0.50	0.65	0.68	0.91

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S = Electric SOFC + all thermal reject heat utilized as steam.

C = Electric SOFC + thermal reject heat utilized as steam and for electric generation.

E = All electric system — SOFC + electric generation only from reject heat.

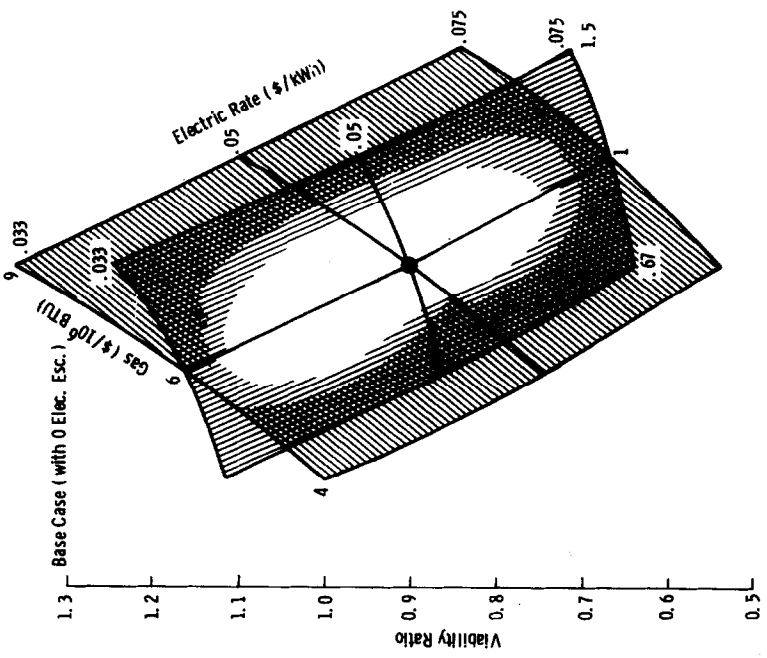
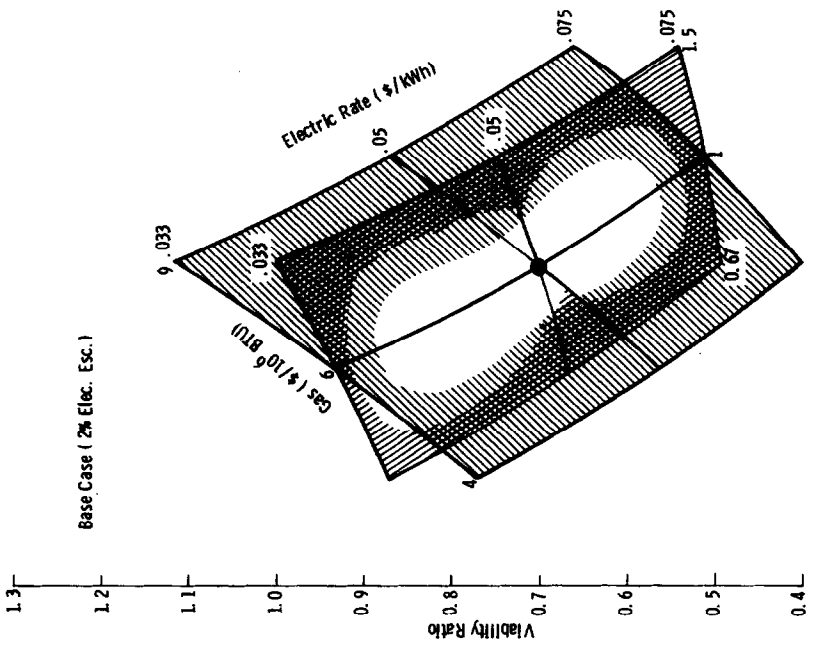


Fig. 10. The effect of electric, gas, and solid oxide fuel cell generator costs on the viability ratio, base case with 2% electric cost escalation.

Fig. 11. The effect of electric, gas, and solid oxide fuel cell generator costs on the viability ratio, base case with 0% electric cost escalation.

displayed good performance and life characteristics under a variety of fuel and operating conditions. A new cell design, lending itself to fabrication into a generator, has been evolved and is well on its way to meeting its expected performance goal. The next phase in advancing this technology toward the ultimate goal is to fabricate a preprototype kW-size SOFC generator.

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