

Clean combined-cycle SOFC power plant — cell modelling and process analysis

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Abstract

The design principle of a specially adapted solid-oxide fuel cell power plant for the production of electricity from hydrocarbons without the emission of greenhouse gases is described. To achieve CO₂ separation in the exhaust stream, it is necessary to burn the unused fuel without directly mixing it with air, which would introduce nitrogen. Therefore, the spent fuel is passed over a bank of oxygen ion conducting tubes very similar in configuration to the electrochemical tubes in the main stack of the fuel cell. In such an SOFC system, pure CO₂ is produced without the need for a special CO₂ separation process. After liquefaction, CO₂ can be re-injected into an underground reservoir. A plant simulation model consists of four main parts, that is, turbo-expansion of natural gas, fuel cell stack, periphery of the stack, and CO₂ recompression. A tubular SOFC concept is preferred. The spent fuel leaving the cell tube bundle is burned with pure oxygen instead of air. The oxygen is separated from the air in an additional small tube bundle of oxygen separation tubes. In this process, mixing of CO₂ and N₂ is avoided, so that liquefaction of CO₂ becomes feasible. As a design tool, a computer model for tubular cells with an air feed tube has been developed based on an existing planar model. Plant simulation indicates the main contributors to power production (tubular SOFC, exhaust air expander) and power consumption (air compressor, oxygen separation). © 2000 Elsevier Science S.A. All rights reserved.

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1. Introduction

Concerns about the impact of anthropogenic CO₂ on global warming have invigorated the search for technologies to reduce the amount of CO₂ that is released to the atmosphere during power generation. CO₂ separation from gaseous mixtures has been applied for many years in the petrochemical industries as well as in natural gas cleaning. Proven technologies are based on physical and chemical absorption. Other technologies, today on a smaller scale, are adsorption processes or membrane gas separation.

Efficiency reductions during CO₂ removal are caused by fuel conversion and pressure losses in the separation equipment. Additional heat and power are needed for solvent regeneration, pressure swing or thermal regenera-

tion, or for compression, in the case of membrane separation. The energy demand of CO₂ separation processes cited in the literature is in the range of 0.026–0.34 kW h/kg CO₂. The corresponding efficiency reduction is in the range of 2.5–11 percentage points depending on the plant type, for example, natural-gas-fired power plants, coal-fired power plants, combined gas and steam cycles.

A unique solution to the problem is offered by SOFCs that are configured to allow recovery of carbon dioxide, which can then be sequestered underground. Fig. 1 shows the basic plant concept. The configuration favoured for the CO₂-separating fuel cell system is based on a tubular design. To achieve CO₂ separation in the exhaust stream, it is necessary to burn the unused fuel without directly mixing it with air, which would introduce nitrogen. To achieve combustion with pure oxygen, the spent fuel is passed over a bank of oxygen ion conducting tubes very similar in configuration to the electrochemical tubes in the main stack of the fuel cell.

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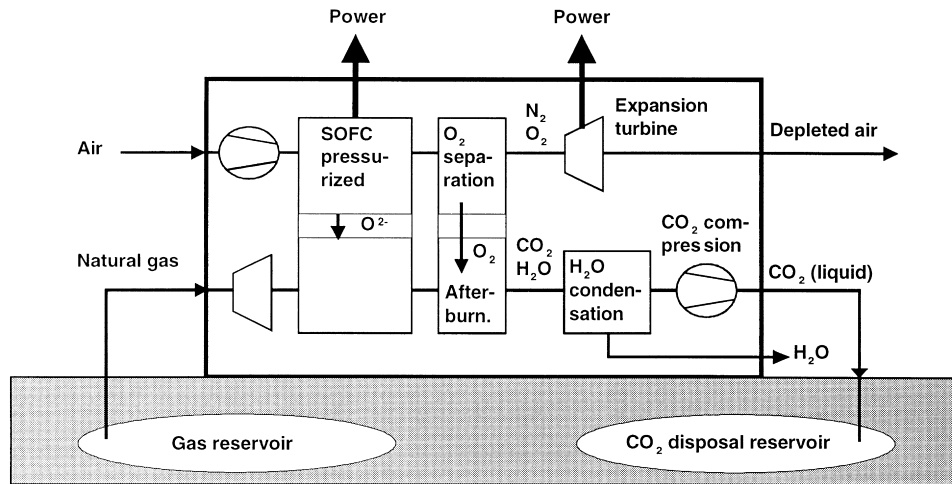


Fig. 1. Emissionless electric power from hydrocarbons.

Optimum conditions for the process involve operating the SOFC under pressure. Exhaust air can still be passed through an expander to recover power as in the conventional SOFC/gas turbine combination. CO₂ has to be brought to high pressure for re-injection and the fuel gas itself may be available at very high pressure from the gas field.

Such permanent underground disposal could take place in depleted gas/oil reservoirs, aquifers or coal seams. Furthermore, injecting CO₂ into productive oil reservoirs can be a means of enhancing oil recovery. Gas production from some coal seams can also be enhanced by CO₂ injection. Schemes for generating power with collection and sequestration of the carbon dioxide produced may suffer from high cost and low efficiency due to the additional CO₂ recovery equipment. In order to assess the economic viability of using SOFCs in this way, simulations of the overall process have been undertaken.

2. CO₂ separation by solid-oxide fuel cells

The configuration favoured for the CO₂-separating fuel cell is based on a tubular design, in which controlled leakage, rather than seals, is used to separate the fuel side from the air side of the cell. This is the design, which has been pioneered by Westinghouse and involves the use of tubular electrodes closed at one end. In the conventional non-CO₂-separating design, exhaust air and exhaust fuel are allowed to mix by controlled leakage of fuel through the baffle boards that separate air and fuel in the cell. By introducing additional baffle boards and careful management of internal flows and pressure drops, it is possible to arrange for the exhaust fuel to be withdrawn as a separate stream without the need to apply high temperature sealants.

Fuel cells do not burn all of their fuel. To keep the electrochemical reaction progressing at reasonable speed

requires a certain partial pressure of unburned fuel to be maintained, resulting in a practical limit to the fuel utilization of 80–90%. To achieve CO₂ separation in the exhaust stream, it is necessary to burn off the unused fuel without directly mixing it with air, which would introduce nitrogen. To do this, as shown in Fig. 1, the spent fuel is passed over a bank of oxygen ion conducting tubes very similar in configuration to the electrochemical tubes in the main stack of the fuel cell.

A flow scheme has been developed for the integrated system of fuel supply, fuel cell, CO₂ recovery and re-injection (Fig. 4). By careful integration of the fuel supply, air supply, and exhaust gas recovery system, the process can be optimized in terms of cost or efficiency. CO₂ has to be brought to a high pressure of at least 200 bar for re-injection and the fuel gas itself may be available at very high pressure from the gas field. Turbo-expansion of the fuel can be used to generate some of the power for CO₂ recompression. The temperature drop created by the expansion process could be used to assist in the liquefaction of the recovered CO₂ so that it can be pumped, rather than compressed, into the reservoir.

Optimum conditions for the process involve operating the SOFC under pressure. Apart from improving the specific power output of the stack, this significantly reduces the compression power for CO₂ recovery and results in smaller CO₂ water separation equipment. Exhaust air can still be passed through an expander to recover power, as in the conventional SOFC/turbine combination, and this form

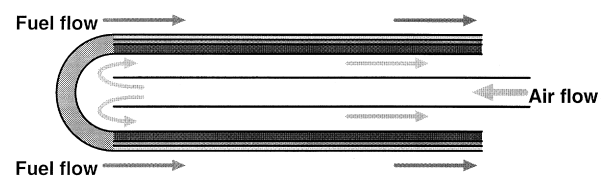


Fig. 2. Tubular SOFC with air feed tube.

Table 1
Data used in the tubular SOFC model

Component	Material	Thickness	Diameter	Length
Air feed tube	Al ₂ O ₃	1.5 m	8 mm	1.5 mm
Cell tube		2.34 mm	22 mm	1.5 m
Air electrode	Doped LaMnO ₃	2.2 mm		
Electrolyte	ZrO ₂ (Y ₂ O ₃)	40 μm		
Fuel electrode	Ni–ZrO ₂ (Y ₂ O ₃)	100 μm		
Interconnection	Doped LaCrO ₃	85 μm		

of integration increases overall efficiency significantly. For simplicity, it is possible to operate the process at atmospheric pressure since the extra compression costs for the CO₂ are not great compared with the overall power output of the fuel cell. They amount to a 2–3% additional loss in overall electrical efficiency. This configuration is, however, not optimum with regard to either cost or efficiency.

3. Modelling of the tubular SOFC

3.1. Adiabatic tubular cell with air feed tube

Based on a planar model [1], an equivalent model was developed for an adiabatic tubular AES (air electrode supported) cell [2] with an air feed tube. The arrangement of the tubes is shown in Fig. 2. The anode gas flows outside the tube walls in co-flow with the cathode gas. The materials and the geometrical data used in the tubular model are given in Table 1.

The equations of an analytical solution given by Nisanoglu [3] were used to calculate the ohmic resistance. Parametric studies show that the long current path in the anode and cathode, along the circumference of the tube, results in an additional voltage drop of the order of 100 mV for typical conditions. This disadvantage of the tubular concept can be compensated by pressurized operation as

shown in Fig. 3. Three curves are shown for pressure levels of 1, 5, and 10 bar, respectively. The mean fuel temperatures are in a range between about 950°C (300 mA/cm²) and about 1000°C (600 mA/cm²). The simulated performance characteristics for pressures of up to 10 bar are in good agreement with experimental data published by Singhal [2]. In comparison to atmospheric operation, the simulated cell voltage is about 60 mV higher for a pressure level of 5 bar and about 100 mV higher for a pressure of 10 bar. The reason for this difference is an increase of the Nernst voltage caused by the higher partial pressure of hydrogen. The current–voltage curve for a pressure of 5 bar shows that a cell voltage of 0.7 V leads to a relatively high mean current density of about 300 mA/cm².

3.2. Tubular cells with directly heat-integrated stack reformer

In the Westinghouse 25 kW generator module, the reformer was appended to the stack [4]. The reformer, which converted 75% of the natural gas, was heated by the exhaust gas. Thus, the cell tubes were in an adiabatic operational mode.

In the new design for the 100 kW SOFC module, the reforming process is, to a large extent, directly heat-integrated with the electrochemical oxidation in the tubular cells [5]. In a first reforming step, the higher hydrocarbons and a small amount of methane are converted in an adiabatic pre-reformer located in the anode gas recycle loop. In a second reforming step, the methane is completely converted in a stack reformer arrangement.

The prototype cells are arranged in 3 × 8 cell bundles, and the bundles into bundle rows. A stack reformer is placed between each bundle row. The reformers are radiantly heated by the adjacent rows of cells. Therefore, the corresponding fraction of the electrochemical excess heat

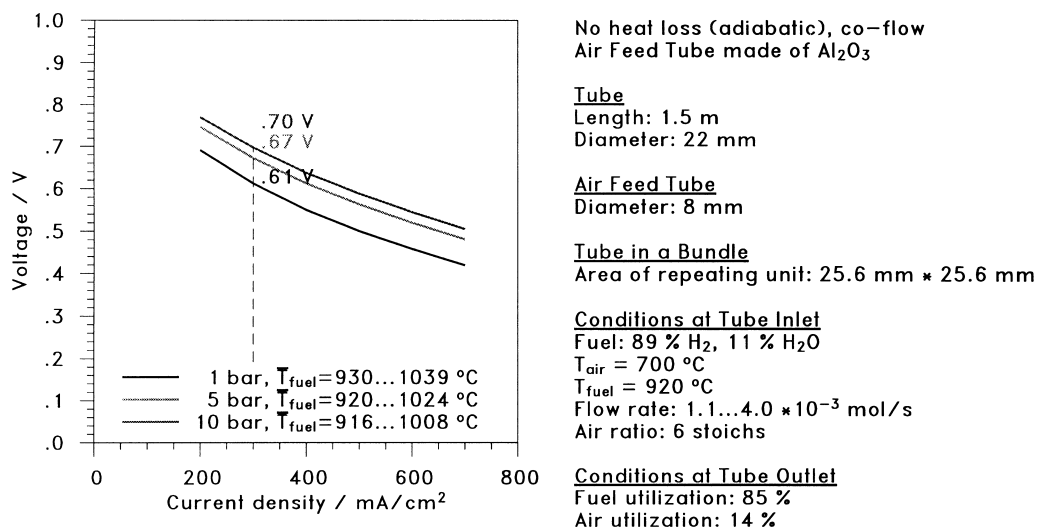


Fig. 3. Effect of pressure on the current–voltage curve.

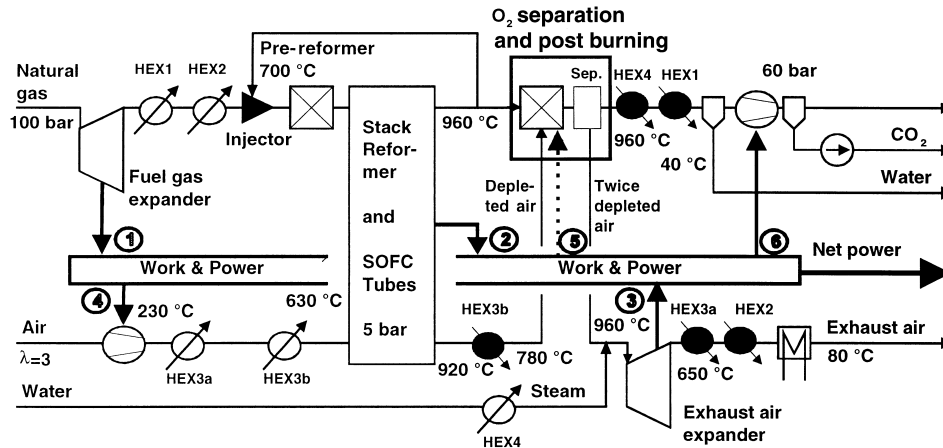


Fig. 4. Simulation model of a 15-MW emissionless SOFC plant.

is directly transferred to the reforming process and is not transported via the cooling air.

Previously, such a radiative heat transfer had not been invoked in the tubular model. Therefore, the module was simulated by an approximation process. The stack reformer was simulated by an equilibrium reactor with a specified methane conversion of 100%. The heat required for gas heating and the reforming reaction was calculated. The extraction of this reforming heat from the tubes was simulated by introducing an additional cooling air stream (not shown in Fig. 4). The amount of this additional air stream was adjusted in such a way that in a subsequent heat exchanger, exactly this amount of reforming heat was extracted over a temperature range equal to the temperature increase in the cell tubes, thus achieving a correct heat balance.

4. Simulation of the CO₂-separating SOFC plant

For an energy analysis of the SOFC power plant concept, the commercial flow sheet simulator PRO/II (Simsci)

was used. This program simulates the components' mass flows and conditions and calculates the energy demand or energy production of common peripheral units. The SOFC stack modelling program is integrated as a FORTRAN subroutine. This is a proven program system for the sensitivity analysis and optimization of SOFC systems [6,7].

In Fig. 4, the components of the plant simulation model are shown. The natural gas stream was expanded and recuperatively heated before entering the SOFC module. Within an anode gas recycle loop, which was accomplished by an injector, the pre-reformer and the stack reformer converted the fuel to a hydrogen-rich gas. The fresh air was compressed to about 5 bar and recuperatively pre-heated to about 600°C. The spent fuel was burnt with pure oxygen. This oxygen was produced in oxygen separation tubes.

Fig. 5 illustrates the basic function of this unit. Heat was produced from the electric power used for improved oxygen separation and by the combustion of the spent fuel with oxygen. In order to limit the temperature of the burnt fuel and the twice-depleted air to about 1000°C, the inlet

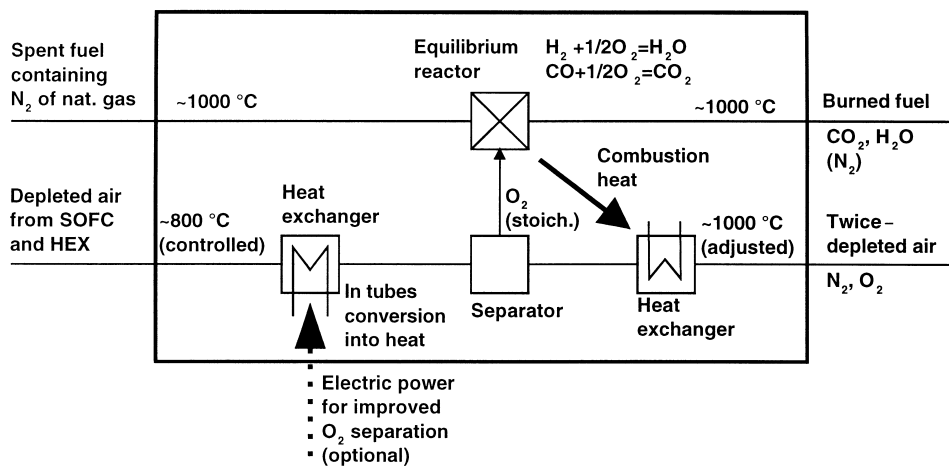


Fig. 5. Simulation model for oxygen separation and spent fuel burning.

temperature of the depleted air coming from the SOFC was tightly controlled. In the recuperative heat exchanger, HEX3b, the depleted air was cooled from 920° to 780°. To avoid an expensive external high temperature heat exchanger, the module had to be designed in such a way that heat was exchanged within the module, as adopted in previous Westinghouse equipment. Additionally, a limited stream of fresh air may be bled into the hot, depleted air in order to adjust the required inlet temperature of the oxygen separation unit.

The pressurized depleted air was expanded in a turbine to produce work, used for air compression, and AC power in a generator. The completely converted anode flue gas was cooled down to separate the water. Then, the CO₂-rich gas was compressed to about 60 bar for liquefaction. In this liquid phase, CO₂ can easily be pumped even to higher pressures. In Fig. 4, a reservoir of work and power is shown. The incoming and outgoing energy flows describe the energy balance for the integrated system. The energy produced in the SOFC, the fuel gas expander, and the exhaust air expander contribute positively to the net power, whereas the energy demand for air compression, O₂ separation, and CO₂ compression lower the net power production.

5. Simulation results

First simulation results and estimations indicate the main parameters influencing the overall net system efficiency. The main contributor to power production is the tubular SOFC module. Depending on the pressure level (5–8 bar) and the chosen current density (200–300 mA/cm²), a cell voltage in the range of 650–750 mV is achieved. This results in a gross electric efficiency of about 50–60% for a fuel utilization of 85%. The main contributors to power consumption are air compression, recompression of recovered CO₂, and the conversion of DC to AC power.

Additionally, depending on the development status of different types of oxygen separation tubes and the special design with respect to oxygen flux densities, a more or less high proportion of DC power is required for this process step. For every 0.1 V applied to the oxygen separation tubes, the plant efficiency decreases by about 1.5% for 85% fuel utilization; 15% of the incoming fuel is burnt with pure oxygen.

Fig. 4 shows in detail the system components and the operation parameters for the reference case. The reference case is defined by the input data for the simulation given in Table 2. The energy data given in the following and in Table 3 are related to a normalized input of natural gas of 100 kW with respect to the lower heating value (LHV). It is assumed that the natural gas has a pressure of 100 bar. The fuel is expanded in two steps. The power, which can

Table 2

Reference data of the SOFC stack and the peripheral components

<i>Stack data</i>	
Cell voltage	700 mV
Fuel utilization (relative to natural gas feed)	85%
<i>Pressures</i>	
Natural gas	100 bar
SOFC	5 bar
CO ₂ compression	60 bar
<i>Temperatures</i>	
Fuel (pre-reformer inlet)	700°C
Fuel (tube outlet)	960°C
Fuel (afterburning outlet)	960°C
Air (afterburning outlet)	960°C
Water condensation	40°C
Exhaust air	80°C
<i>Flow rates</i>	
Air ratio	3
<i>Conversions in pre-reformer</i>	
Higher hydrocarbons	100%
Methane	10%
<i>Conversion in stack reformer</i>	
Methane	100%
<i>Component efficiencies</i>	
Inverter	95%
Air compressor	80%
CO ₂ compressor	80%
Exhaust air expander	70%

be produced in this unit, is relatively low. The contribution to power production is only 0.5 kW per 100 kW(LHV) natural gas. The gas is cooled to –54°C and –50°C, respectively. Then, it is recuperatively heated in two steps to about 150°C before entering the injector in the anode gas recycled loop.

By mixing the natural gas with the recycled anode gas, a temperature of 700°C can be set at the inlet of the adiabatic pre-reformer. Complete conversion of the higher hydrocarbons and 10% conversion of the methane results in a temperature decrease to 530°C. In the stack reformer, the pre-reformed gas is heated and the methane is completely converted before entering the SOFC tubes. The total heat consumption of the stack reformer is 35 kW; the reforming heat without pre-heating amounts to 24 kW. The fuel temperature at the cell outlet is adjusted to 960°C. A major fraction of the gas is recycled. The other fraction enters the oxygen separation tubes for burning with oxygen.

In the SOFC tubes, the cell voltage is adjusted to 700 mV and the fuel utilization to 85% by selecting 383 tubes per 100 kW(LHV) natural gas. A current density of 255 mA/cm² is calculated and 57 kW of DC power is produced.

Table 3
Results of simulation — energy balance (normalized values)

		DC power consumption of oxygen separation		
		Zero	Medium	High (1 V)
<i>Input natural gas (LHV), normalized</i>	kW	100	100	100
<i>Stack data</i>				
Fuel utilization	%	85	85	85
Cell voltage	mV	700	700	700
Current density	mA/cm ²	255	255	255
<i>Power production</i>				
AC power from fuel gas expander (1)	kW	0.5	0.5	0.5
AC power from SOFC (2)	kW	54	54	54
Work from exhaust air expander (3)	kW	32	32	32
AC power from expansion/air compression (3 + 4)	kW	11	11	11
<i>Power consumption</i>				
Work for air compression (4)	kW	–21	–21	–21
DC power for oxygen separation (5)	kW	–	–5 to –10	–15
AC power for CO ₂ compression (6)	kW	–1.0	–1.0	–1.0
<i>Net electrical plant efficiency</i>	%	64	54–59	49

The amount of cooling air corresponds to a stoichiometric air ratio of 3. Such a relatively low value is possible because a high amount of heat produced in the cell tubes is directly transported to the stack reformer. Additionally, the air feed tube leads to a high air temperature increase in the stack which directly lowers the amount of air required for limiting the temperature increase of the cell tube material. The fresh air is compressed to about 5 bar, which requires 21 kW. The air is recuperatively heated from 230°C to 630°C. In the air feed tube and in the cell tube, an air temperature increase of 290 K is calculated. The air temperature at the cell outlet is 920°C. The depleted air is cooled to 780°C in an air recuperator. The exhaust outlet temperature, which is adjusted to 960°C, is controlled by this air temperature.

The twice-depleted air is expanded in a turbine. Here, the temperature decreases from 960°C to 650°C, while 32 kW of work is produced. The hot exhaust air is recuperatively cooled to 280°C. Between this temperature and 80°C, 21 kW useful heat can be extracted.

The hot burnt fuel leaving the oxygen separation tubes is recuperatively cooled to 40°C. The condensed water is separated and the remaining CO₂ is compressed to 60 bar for liquefaction.

The performance data of the tubular cells and the energy balance for the complete system are listed in Table 3. The main contributors to power production are the tubular SOFC (57 kW DC) and the exhaust air expander (32 kW). A great deal of power is consumed by the air compressor (21 kW). The oxygen separation needs to be designed in such a way that the DC power consumption is limited to an acceptable value of 5–10 kW.

The net system efficiency is of the order of 55–60% for this reference case, at 5 bar, 700 mV. Here, it is assumed

that the number of oxygen separation tubes limits the corresponding power consumption with a loss of system efficiency 5–10%.

6. Plans for demonstration

The present plans for developing this technology focus on a demonstration of the CO₂-separating fuel cell at the 100-kW level. SOFC technology is modular and this scale is considered sufficient to verify the concept for any power level. The demonstrator will not re-inject the recovered CO₂, as the quantities produced by a 100-kW unit during the test are too small to justify drilling an injection well. Subsequent to a successful test, consideration will be given to scaling up the technology to achieve a power output of several tens of MW, probably in an oil field application.

7. Conclusions

The design principle of a CO₂-separating SOFC system for the production of electricity from hydrocarbons without the emission of greenhouse gases is described. First energy analyses and estimations indicate the main contributors to power production (tubular SOFC, exhaust air expander) and power consumption (air compressor, oxygen separation). Depending on the pressure level (5–8 bar) and the current density chosen (200–300 mA/cm²), a cell voltage in the range of 650–750 mV is achieved. For a fuel utilization of 85%, this results in a gross electric efficiency of about 50–60% for the tubular SOFC and 60–70% for the SOFC/turbine combination.

The net system efficiency is of the order of 55–60% for the reference case at 5 bar, 700 mV. Here, it is assumed that the number of oxygen separation tubes limits the corresponding power consumption with a loss of system efficiency 5–10%.

The CO₂-separating fuel cell system described represents an example of an application in which the fuel cell can provide functionality that other generation systems will find difficult to match. For a breakthrough into the marketplace, further work has to be done to optimize the flowsheet, the design of the key components, and the operating parameters (fuel utilization, pressure, and temperature levels) with appropriate consideration given to energetic and economic aspects.

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