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## MODELING OF A METHANE FUELLED DIRECT CARBON FUEL CELL SYSTEM

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## ABSTRACT

Energy conversion today is subject to high thermodynamic losses. About 50 to 90 % of the exergy of primary fuels is lost during conversion into power or heat. The fast increasing world energy demand makes a further increase of conversion efficiencies inevitable. The substantial thermodynamic losses (exergy losses of 20 to 30 %) of thermal fuel conversion will limit future improvements of power plant efficiencies. Electrochemical conversion of fuel enables fuel conversion with minimum losses. Various fuel cell systems have been investigated at the Delft University of Technology during the past twenty years. It appeared that exergy analyses can be very helpful in understanding the extent and causes of thermodynamic losses in fuel cell systems. More than 50 % of the losses in high temperature fuel cell (MCFC and SOFC) systems can be caused by heat transfer. Therefore system optimisation must focus on reducing the need for heat transfer as well as improving the conditions for the unavoidable heat transfer. Various options for reducing the need for heat transfer are discussed in this paper. High temperature fuel cells, eventually integrated into gas turbine processes, can replace the combustion process in future power plants. High temperature fuel cells will be necessary to obtain conversion efficiencies up to 80 % in case of large scale electricity production in the future. The introduction of fuel cells is considered to be a first step in the integration of electrochemical conversion in future energy conversion systems.

*Keywords:* Fuel cell systems; Exergy analysis; Thermodynamic analysis; System modelling; Cycle Tempo; PEMFC; MCFC; SOFC; Applications

### INTRODUCTION

Today our world strongly depends on the availability of energy for almost all of their activities. Total energy demand is growing fast, in particular due to the development of the large Asian countries. To ensure our future energy supply the losses of energy conversion have to be reduced and the utilisation of available sources, in particular renewable sources, should be stimulated. Conversion of primary fuels into electricity, power, heat or secondary fuels is necessary to fulfil our energy demands. These conversions are involved with substantial thermodynamic losses. These losses should be presented as exergy losses to get a true representation of the thermodynamic performance of conversion systems. Exergy is the potential to obtain work from an amount of energy or from an energy flow; exergy values do represent a true yardstick for all relevant thermodynamic characteristics [1].

Exergy efficiencies are some percentage points lower than thermal efficiencies (based on the lower heating value of the fuel) in case of the conversion of primary energy into power. As thermal efficiencies of less than 50 % are normal practice exergy losses will on average be higher than 50 %. Thermal efficiencies are in general high (around 90 % or more) in case of heat generating systems, but exergy efficiencies are much lower depending on the temperature level at which is transferred to the process or transport fluid. In case of hot water boilers for space heating exergy losses are 85 to 90 %. These numbers show that there is still a huge potential for exergy loss reductions. This paper will discuss the possibilities to reduce these losses by applying electrochemical conversion processes. Furthermore several options are mentioned that will improve the thermodynamic performance fuel cell systems.

# LIMITATIONS OF CONVENTIONAL ENERGY CONVERSION SYSTEMS

Conversion efficiencies of energy systems depend on the size of the system. The highest efficiencies are obtained for systems that deliver some hundreds megawatts of electricity or more. Figure 1 shows the exergy flow diagram (Grassmann diagram) of a 600 MW power plant with a steam turbine cycle. The diagram shows that the main thermodynamic losses (exergy losses) of this plant occur in the boiler. The fuel supply to the plant represents an exergy flow of 1471 MW from which 382 MW is lost during combustion. This means that more than 25 % of the supplied exergy is lost due to the conversion of fuel into heat. In [2] it is shown that for atmospheric combustion the exergy loss will always be more than 20 % even when very high air preheating temperatures are applied. Heat transfer in the boiler, from flue gas to steam cycle, causes an additional exergy loss of 315 MW. Further losses in the steam cycle and auxiliary equipment are small in comparison with the losses in the boiler. The losses caused by heat transfer can be reduced by increasing the steam pressure and temperature at turbine inlet or the application of topping cycles. Topping cycles with MHD generators as well as mercury and potassium turbines have been investigated. However only gas turbine topping cycles in socalled combined cycles are successfully applied for natural gas fired plants. But unfortunately the exergy loss of combustion remains almost unaffected. Figure 2 shows the exergy flow diagram of a 379 MW combined cycle plant with triple pressure heat recovery steam generator. The results are derived for a modern gas turbine. Gas turbines based on the most advanced technology (steam cooled blades) however will show somewhat better results. The exergy loss of combustion is 200

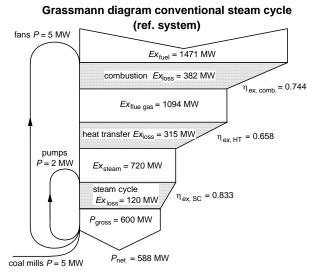
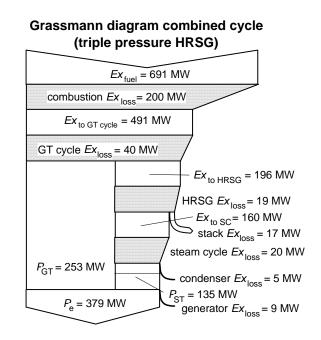


Figure 1. Exergy flow diagram (Grassmann diagram) of a 600 MW conventional steam turbine plant



## Figure 2. Exergy flow diagram (Grassmann diagram) of a 379 MW combined cycle plant

MW which is almost 29 % of the exergy of the supplied fuel. Gas turbines with steam cooled blades will show somewhat lower combustion losses but it is not expected that substantial reductions can be realised.

Reflections with regard to feasibility of further improvements of the efficiency of thermal power plants can be clarified also with the following equation for the thermal efficiency:

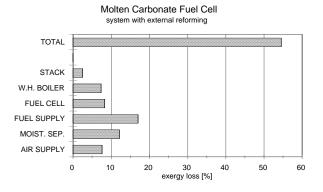
$$\eta_{\text{th, plant}} = \eta_{ex, \text{ intern}} \cdot \left( 1 - \frac{\overline{T}_{\text{C}}}{\overline{T}_{\text{H}}} \right)$$

In this equation the so-called Carnot factor  $\left(1 - \frac{\overline{T}_{C}}{\overline{T}_{H}}\right)$  is

multiplied with an efficiency,  $\eta_{ex, \text{ intern}}$ , that represents the internal losses of the thermal power cycle. The Carnot factor is based here on the thermodynamic equivalent temperatures of heat transfer to the cycle  $(\overline{T}_{\rm H})$  and from the cycle  $(\overline{T}_{\rm C})$ . Actually all thermodynamic losses due to combustion and heat transfer to and from the cycle are included in this factor. Losses due to irreversibility's within the cycle are included in the internal efficiency of the cycle. For the considered combined cycle plant the overall Carnot factor is about 72 % and the internal efficiency 80 %. Raising the Carnot factor till 80 % will require an increase of the thermodynamic equivalent temperature of heat transfer to the gas turbine cycle of 400 K. It is very unlikely that such an increase can be realised within one or two decades. However when it is assumed that an increase of the Carnot factor to 80 % is possible and that the internal efficiency can be improved to 90 %, the overall thermal efficiency of a combined cycle plant will become 72 %. Therefore it may be stated that plant efficiencies over 80 % can not be realised without introducing more efficient fuel conversion systems.

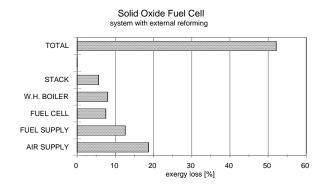
### CHARACTERISTICS OF FUEL CELL SYSTEMS

An extensive evaluation of exergy losses in high temperature fuel cell systems is presented in [3]. For an arbitrarily chosen Molten Carbonate Fuel Cell system (power level around 1 MW) with external reforming the exergy losses are presented in figure 3. The calculated exergy losses are allocated to a limited number of subsystems like flue gas exhaust (stack), waste heat boiler (w.h. boiler), fuel cell stack



### Figure 3. Exergy losses in a Molten Carbonate Fuel Cell system with external reforming

(fuel cell), fuel supply system including the reformer, moisture separation and air supply system. Without going into details, it may be concluded from this figure that the exergy losses of the fuel cell stack, which are actually the losses of electrochemical fuel conversion, are small in comparison with the losses of thermal combustion. As rather conservative input data are applied for the cell performance a further reduction of the fuel cell exergy losses can be expected. Similar results are found for a Solid Oxide Fuel Cell system with external reforming. The exergy losses of this system are shown in figure 4.



## Figure 4. Exergy losses in a Solid Oxide Fuel Cell system with external reforming

The overall electrical efficiencies of both systems are rather low (about 46 and 48 %). The system designs are established mainly for evaluating the effect of specific parameters and are therefore not fully optimised. More detailed insight into the losses in fuel cell systems can be obtained by distinguishing also the causes of exergy loss in all subsystems as shown in figure 5 for the same MCFC system as in figure 3. From figure 5 it appears that the majority of exergy losses is caused by heat transfer in the system. Losses in the fuel cell are mainly due to charge transfer caused by the equivalent cell

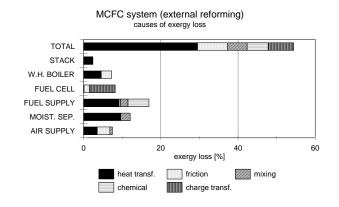


Figure 5. Causes of exergy losses in a Molten Carbonate Fuel Cell system with external reforming

resistance. A decrease of the equivalent cell resistance will not reduce only the exergy losses in the cell but will also limit the need for heat transfer and consequently the extend of exergy losses due to heat transfer. It may be concluded from this evaluation that electrochemical conversion is beneficial because of the much lower exergy losses of the combustion process and also that heat transfer is the main reason for exergy losses in fuel cell systems. Therefore when designing fuel cell systems various measures have to be considered in order to limit the exergy losses due to heat transfer.

The total loss of fuel cell stacks and fuel supply should be compared with the exergy losses of combustion for a true comparison with thermal combustion systems. System studies have demonstrated that also the decrease of losses in the fuel supply system is conceivable.

## APPLIED SOFTWARE FOR EVALUATING FUEL CELL SYSTEMS

Before discussing the effect of various design options of fuel cell systems some remarks will be made on the software used for the reported evaluations.

At the Delft University of Technology (TU Delft) it was concluded that the integration of specific models for fuel cell systems, like models for fuel cells and reformers, into an existing software package, Cycle-Tempo [4], for the thermodynamic evaluation of energy systems will be useful. From the beginning it was clear that this approach has the disadvantage that apparatus models in this type of software must be relatively simple. But it enables the evaluation of integrated energy systems with fuel cells and conventional energy conversion equipment. Furthermore, as Cycle-Tempo is provided with extensive facilities for the presentation and analysis of the results of system calculations, these facilities will become available also for the evaluation of fuel cell systems.

Cycle-Tempo has been extended with fuel cell models for all regular considered fuel cells like the alkaline fuel cell (AFC), phosphoric acid fuel cell (PAFC), solid polymer fuel cell (SPFC) or polymer electrolyte membrane fuel cell (PEMFC), molten carbonate fuel (MCFC), solid oxide fuel cell (SOFC) and direct carbon fuel cell (DCFC). Models with direct and indirect internal reforming are included for the high temperature fuel cells, the MCFC and SOFC. The SPFC (PEMFC), MCFC and SOFC models have been made for design and off-design calculations. In design mode desired performance factors as cell voltage and current density can be specified. For off-design the voltage-current density relation must be defined. In case of high temperature fuel cells the following simple equation is applied:

$$V_{\text{cell}} = V_{\text{rev}}(u) - i(u) \cdot R_{\text{eq}}$$

This equation indicates that the local current density, i(u), in de fuel cell is determined by the difference between the actual cell voltage,  $V_{cell}$ , the reversible cell voltage,  $V_{rev}(u)$ , and the equivalent cell resistance,  $R_{eq}$ . The voltage-current density relation for the SPFC (PEMFC) is based on the Tafel equation:

$$V_{\text{cell}} = V_{\text{rev}} - (i + i_{\text{n}}) \cdot R_{\text{eq}} - A \cdot \ln\left(\frac{i + i_{\text{n}}}{i_{0}}\right) + B \cdot \ln\left(1 - \frac{i + i_{\text{n}}}{i_{1}}\right)$$

In this equation is  $i_n$  the internal and fuel crossover equivalent current density, A the slope of the Tafel line,  $i_0$  the exchange current density at the cathode, B the constant in the mass transfer over-voltage equation and  $i_1$  the limiting current density. All models have been developed as one-dimensional isothermal models. Investigations to check the accuracy of these conditions for high temperature fuel cells ([5] and [6]) have indicated that good correspondence with measured fuel cell performance data can be achieved.

Because of the universal layout of the fuel cell models, extensions with new types of fuel cells are rather easily possible. Recent investigations into the innovative MCFC (i-MCFC) have resulted in a preliminary model for this type of fuel cell.

## **EVALUATION OF VARIOUS DESIGN OPTIONS**

Since the start of the fuel cell system studies at the TU Delft, about twenty years ago, a large number of fuel cell systems are evaluated. During this period several system studies of conventional alternatives have been made in parallel. Based on these experiences some universal conclusions can be drawn and specific design options and applications will be discussed. Most of the fuel cell system evaluations are concerned with high temperature fuel cells (MCFC and SOFC) Only a limited number of PEMFC systems are investigated.

#### PEMFC systems

The development of PEMFC systems is strongly focusing on transport applications. This development is supposed to be beneficial also for small stationary systems like micro-CHP. However the boundary conditions for micro-CHP systems are not clear, as there is not really an existing market today. Micro-CHP systems based on a small gas engines, with electrical efficiencies of 27 % (LHV) and overall thermal efficiencies of 90 % (LHV), are commercially available. But present market conditions are not stimulating the replacement of central heating boilers by such systems. The higher capital costs can only be compensated when reasonable yearly load factors are achieved. Load factors of heating systems are limited by climate conditions. And favourable conditions for the delivery of electricity into the grid are necessary to enable almost continues operation during the cold season. Micro-CHP systems based on PEMFC's can provide higher electrical efficiencies and lower emission levels than the existing natural gas fuelled micro-CHP unit. But capital costs should be low. Performance and costs of natural gas fuelled PEMFC CHP systems are mainly determined by the applied fuel cell parameters and fuel conversion process. Possibilities for heat integration are limited because of the large temperature difference between the fuel cell and the fuel conversion process. After all it is not possible to use low temperature heat from the fuel cell stack for the high temperature fuel conversion process.

Steam reforming (SR), partial oxidation and auto-thermal reforming (ATR) are considered for the conversion of natural gas into hydrogen. Steam reforming appears to be the preferred process (see [5], [6], [7], [8]) for this application because of the high hydrogen yields and concentrations, the high methane conversion rate and the relatively low CO concentration. The costs of a fuel processor based on steam reforming are estimated to be less than 350 \$/kWe in case of mass production [8]. Electrical efficiencies of PEMFC units with steam reformer, assuming state of the art technology, are in the range from 28 [7] to 31-38 % [6]. Units with auto-thermal reforming have somewhat lower efficiencies: 24-33 % [6]. The same reference gives expected values for the future: 41 % for units with ATR and 45 % for units with SR. The performance of micro-CHP systems must be compared with usual way of energy supply: the separate generation of electricity, in large central power plants, and heat, in central heating boilers. Electrical efficiencies higher than 35 % and overall efficiencies exceeding 90 % are supposed to be necessary when micro-CHP units must compete with large combined cycle plants. Micro-CHP units can only be beneficial with respect to fuel consumption if the exergy efficiency of the unit is higher than the exergy efficiency of separate generation of electricity and heat. In island operation (without delivery to the grid) the exergy efficiency is determined only by the heat to power ratio. That means that the exergy efficiency of a CHP plant can be increase only by increasing the demanded power to heat ratio and can not be influenced further by the design of the CHP unit. Discussions about the optimum power to heat ratio become more complicated if delivery of electricity to the grid is possible. Then the attractiveness of micro CHP is often dominated by the prices for imported and exported electricity. The further increase of electrical efficiencies of central power stations will favour the application of electrical driven heat pumps. The application of micro-CHP units therefore will be

#### Solid Polymer Fuel Cell micro CHP system

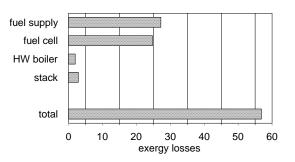


Figure 6. Exergy losses in a micro CHP system based on Solid Polymer Fuel Cells (PEMFC) and ATR

questionable for the long term [9].

The exergy losses in a modelled micro-CHP unit with PEM fuel cells and fuel processing with auto-thermal reforming are shown in figure 6. The calculated net electrical efficiency of the unit is 38.7 %, based on the LHV of the fuel, and the overall thermal efficiency 91.1 %. The corresponding exergy efficiencies are 37.3 and 43.1 %. Figure 6 shows that the majority of the exergy losses arise in the fuel cell stack and the fuel supply system. The remaining losses, due to heat transfer from flue gas to the hot water (HW boiler) and the discharge of cooled flue gas to the atmosphere (stack), contribute only marginally to the overall exergy loss of the unit. In comparison with the MCFC and SOFC systems the exergy losses in the fuel cell are very large. It is expected however that these losses can be reduced in future plants. Losses in the fuel cell result mainly from the differences between the actual cell voltage and the reversible cell voltage. Further reductions of cell resistance's will of course decrease the exergy loss of the cell. The nominal cell voltage, which was supposed to be 0.7326 V, also determines the amount of heat generated in the fuel cell stack. Because of the low temperature of the fuel cells, heat is discharged at low temperature. And consequently this heat has a low specific exergy. The introduction of high temperature polymer fuel cells [10] can increase the temperature of the discharged heat and therefore reduce the exergy losses of the fuel cell stack. However this will improve the overall system performance only as this heat is actually used at higher temperatures.

The losses of the fuel supply system are mainly originating from the actual fuel conversion. Losses during shifting and CO removal are relatively small. From [6] it can be concluded that steam reforming results in slightly lower exergy losses than auto-thermal reforming. The effect however is limited. Replacing the thermal conversion of fuel by an electrochemical conversion can provide a more effective way to reduce the losses of the fuel supply system. Replacing the reformer by a SOFC stack with low fuel utilisation can significantly increase the electrical efficiency [11], [12]. Further evaluations of this kind of concepts must clarify whether this solution can be feasible in case of micro-CHP systems.

### MCFC systems

The development of MCFC focuses primarily on small and intermediate scale CHP systems [13] [14], with electrical power levels from roughly 100 kW until 10 MW. Also higher powers are considered. The high temperature of the residual heat enables the generation of steam and thus the application for industrial CHP plants. The market of industrial CHP plants is diverse and it may be expected to be a durable market. For the long term a further integration of the production of electricity and industrial products might provide new challenges for HT fuel cell systems.



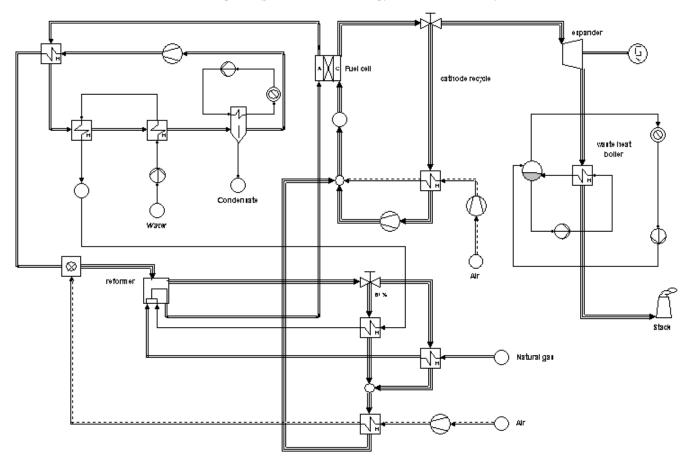
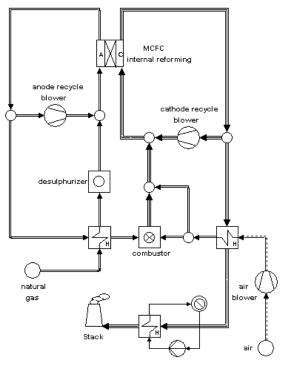


Figure 7. Pressurised MCFC-CHP system with external reformer

It is concluded before that heat transfer is the main cause of exergy loss in HT fuel cell systems. This conclusion corresponds with the results of initial exploring system studies [15]. It appeared that internal reforming and recycling of anode and cathode outlet streams will raise the electrical efficiency seriously. It will be clear that internal reforming reduces the need for heat transfer. Actually heat is transferred within the stack, but as the difference between the temperature of the fuel cell and the equilibrium temperature of the reforming reaction is limited and all temperatures are high the exergy loss due to this heat transfer can be largely reduced. In general the cathode flow is used to discharge the majority of the generated heat from a MCFC stack. As the temperature rise of the flow in the

#### atmosferic MCFC-CHP with internal reforming



## Figure 8. Atmospheric MCFC-CHP plant with internal reforming

stack is limited to approximately 100 K, a large cathode flow is necessary. Recycling the cathode flow (see figure 7) will reduce the flow of fresh air that must be heated as well as the flow of hot off-gas that must be cooled. In figure 7 the purpose of the heater in the recycle flow is only to decrease the operation temperature of the recycle blower. In MCFC systems the anode off-gas flow can be recycled to the cathode inlet in order to provide the necessary carbon dioxide. The unconverted fuel must be oxidised first which occurs in the combustor of the reformer. Before combustion water vapour is removed. The removal of water vapour results in a higher combustion temperature in the reformer and higher concentrations of carbon dioxide at the cathode. Under atmospheric conditions the condensation of water vapour occurs at low temperature (< 100 °C). The removal of most of the available water vapour requires the extraction of an substantial amount of heat. Commercial application of heat at these low temperatures will be difficult. Increasing system pressure can increase the condensation temperature. A system pressure of 4 bar enables heat extraction during condensation for the generation of hot water to be used for instance for district heating. This will increase the overall thermal efficiency of the plant with almost 20 % and it enables the generation of low temperature heat (hot water) without large exergy losses. A further effect of elevated system pressure is the application of an expander for the generation of electricity. It reduces the off-gas temperature before the gas enters the waste heat boiler. As the temperature of industrial steam is in general not more than 200 °C the large temperature difference between gas and steam in the waste heat boiler will cause significant exergy losses. Therefore the expander will allow for a more efficient use of exergy from the off-gas flow. If the generation of steam is not useful the remaining heat from the off-gasses can be used for additional power production. In case of small-scale systems the application of Organic Rankine Cycles (ORC's) can be beneficial. For maximum power generation the ORC working fluid should be adapted to the off-gas temperature [16].

In the system flow diagram of figure 7 reformer steam is generated in a series of heaters. Since the anode off-gas contains large amounts of water vapour recycling of anode offgas to the reformer can eliminate the generation of reformer steam. In case of internal reforming this will significantly simplify the system layout, as will be clear from figure 8, and will reduce the total surface area and costs for heat transfer. This solution enables also the reduction of exergy losses of heat transfer in the system. Fuel Cell Energy Inc. (formerly ERC) has promoted this solution for many years; however recent system designs are provided with separate steam generators.

Some further options to reduce the heat generation in fuel cell stacks have been investigated. It was suggested in the past that the cell geometry (flat plate, circular or tubular) could affect the current density distribution and consequently the losses in the cell. In [17] however it was demonstrated that in case of fuel cells with external reforming the geometry has no or only a negligible effect on the current density. In this investigation cells with internal heat transfer were not included; therefore no conclusions are drawn regarding cells with internal reforming. An investigation of the effect of co-flow versus counter-flow of anode and cathode flows [17] has demonstrated that co-flow has slight advantages over counter-flow. Assuming equal average current densities co-flow offers a relatively homogeneous current densities.

The concentration of hydrogen at the anode of an MCFC will change as a function of fuel utilisation because of the

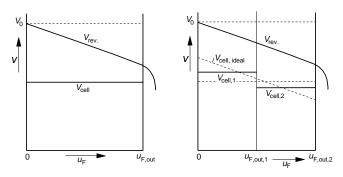


Figure 9. Cell voltages in case of single stage (left) and two stage (right) operation

consumption of hydrogen and the addition of reaction products (water vapour and carbon dioxide); the concentration of oxygen reduces also while the concentration of water vapour increases. Therefore the reversible cell voltage in high temperature fuel cells decreases as a function of fuel utilisation, which causes an non-homogeneous current distribution and relatively large exergy losses. Multistage oxidation, a series connection of fuel cell stacks (with regard to the gas flows), provides an option to reduce the differences in current density within the cell. Figure 9 shows in principle the effect of two stage oxidation versus single stage oxidation: two stage oxidation decreases the difference between the reversible voltage and the actual cell voltage at cell inlet  $(u_{\rm F} = 0)$  and increases the voltage difference at cell outlet. Thus a more uniform current density can be derived. Assuming the same average current density, system calculations have demonstrated a 1 % (point) higher electrical efficiency of for two stage oxidation [17]. The additional effect of 3 or more stages is limited; in case of an infinite number of stages, in figure 9 reflected as  $V_{\text{cell, ideal}}$ , the efficiency increase over the single stage case will be about 1.33 %. The calculations made to demonstrate the effect of multistage oxidation were initially based on the assumption that the cell resistance is constant, also when the operation temperature of the cell changes. Afterwards, when relations between cell temperature and resistance became available, additional system calculations were made to check the influence of changes in cell resistance. It appeared that the increase in efficiency is somewhat lower than initially calculated [18]. On the other hand additional improvements are possible by further optimising the cell temperatures and other design parameters. Multistage oxidation is also possible within one stack as shown in figure 10. As the cells are electrically connected in series each cell will have the same current density. But the voltage of the cells will differ depending on the reversible voltage that results from the fuel composition.

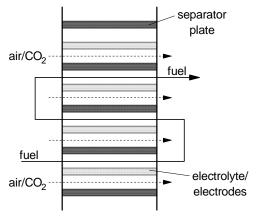


Figure 10. Multistage oxidation in a single stack

The series connection of stacks can also be used for improving the thermal management of the fuel cell system. The cathode gas recycle applied in the system of figure 7 enables the repeated use of cathode gas without fully heating and cooling the cathode flow from and to environmental temperature. As discussed before, this reduces the exergy losses from heat transfer within the system. Recycling gas at high temperatures however requires large powers for driving the

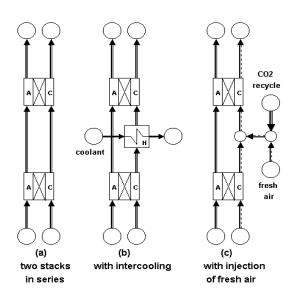


Figure 11. Some options for a series connection of stacks

recycle blower. The application of a series of stacks also enables the reuse of cathode gas and can avoid the need for recycling. Figure 11 option (a) depicts just a series connection of two stacks; the inlet conditions of the second stack are determined by the outlet conditions of the first one. The total temperature rise of the cathode flow is divided over both stacks; the cathode gas flow almost equals the single stack case. Option (b) represents a two stage stack arrangement with intermediate cooling of the cathode flow. In this case the temperature rise of the cathode gas can be maximum in each stack. As the heat capacity of the gas can be used twice the cathode gas flow can be about half the flow of the single stack case. This will result in a significant decrease in capacity of the recycle blower. In case of MCFC stacks heat must be extracted at temperatures of 600 to 700 °C; this heat has a high exergy and should be applied preferably in a bottoming cycle (steam or ORC) or as high temperature process heat. If for instance the power scale does not allow for a bottoming cycle the injection of fresh air in the cathode flow between both stacks (option (c)) can be considered. To balance the oxygen and the carbon dioxide concentrations the anode recycle for the supply of CO<sub>2</sub> must also be divided over the cathode flows to the first and second stack. In case of systems with cathode gas recycle the series connection of stacks can improve the electrical efficiency with more than 5 % (points). An increase from 46 to 55 % has been calculated for a system with four stacks in series and injection of fresh air and anode recycle gas in the cathode flow between the stacks [17].

The operation temperature of MCFC's is generally supposed to be optimum at 650 °C. The availability of empirical equations for the equivalent (or quasi-Ohmic) cell resistance enabled the evaluation of the effect of the average cell temperature on overall system efficiencies [18]. The empirical equations were derived from CRIEPI. Applying these equations it appeared that the cell resistance decreases continuously when increasing the cell temperature from 600 to 700 °C; the cell power shows an optimum around 660 °C. The effect on overall system performance was evaluated for the system design as depicted in figure 7. System calculations have been made for cell temperatures from 600 to 700 °C with temperature steps of 25 °C. The highest electrical efficiency was derived for the system with a cell temperature of 725 °C; also the overall exergy efficiency appeared to be maximum for this case but the overall thermal efficiency was still increasing with temperature. The efficiency curve is rather flat around its optimum. It was concluded that the optimum cell temperature with regard to system efficiency may be not quite the same as with regard to cell efficiency, but differences are nearly negligible.

In MCFC systems the need for rather high concentrations of carbon dioxide at the cathode sometimes complicates the system design. Furthermore mixing air with  $CO_2$  is in conflict with the future need for  $CO_2$  sequestration as discussed today. The concept of an improved MCFC (i-MCFC) might provide a solution for these problems. In the i-MCFC anode recycle gas is not mixed with the cathode air flow but supplied through separate gas channels in the electrolyte matrix [19]. The concept is evaluated and discussed more extensively in [18]. A system calculation has demonstrated that replacing the MCFC by the i-MCFC does hardly affect the system performance. Systems studies that fully exploit the features of the i-MCFC however have not been made.

#### SOFC systems

The development of SOFC systems is mainly focusing on small to intermediate scale power and CHP systems [22] [23] [24]. This focus is partly due to the present state of development; Siemens-Westinghouse [23] is seriously opting for higher powers. Many system studies also are considering SOFC as an appropriate technology for large scale power applications [20] [21]. The integration of SOFC's and gas

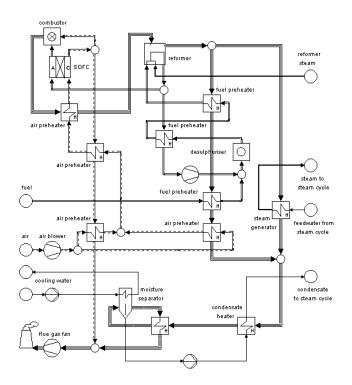


Figure 12. SOFC subsystem at atmospheric pressure with external reforming

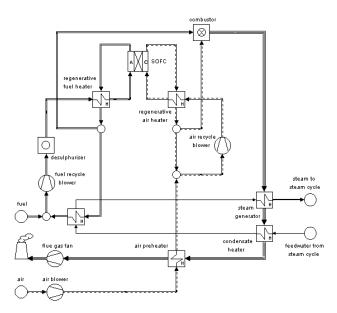
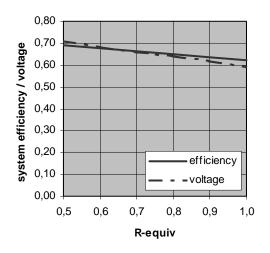


Figure 13. SOFC subsystem with internal reforming and off-gas recycle

turbines appears to provide a very interesting option. SOFC applications show a large overlap with MCFC applications.

Exploring system studies [20] have shown that the system design has a large influence on efficiencies and costs of SOFC plants. Figure 12 shows the layout of a SOFC subsystem with external reforming. Residual heat from this plant is utilised for power production with a steam turbine cycle; the steam turbine cycle however is not included in this figure. From the system calculations it appeared that large amounts of heat must be exchanged internally in this subsystem resulting in large exergy losses and large heat exchanging area's. The economic evaluation has learned that it will be hardly possible to make such a system economically attractive because of the high capital costs. System efficiency as well as economics are improved significantly by applying internal reforming and recycle of anode and cathode off-gas flow. The configuration of such a SOFC subsystem is shown in figure 13. The overall electrical efficiency (including steam turbine power) is increased from 51 % for the case shown in figure 12 to 62 % for the case shown in figure 13, in spite of the lower cell voltage caused by the reduced concentration of hydrogen and carbon monoxide due to anode gas recycle. At the same time the amount of heat transferred within the SOFC subsystem is decreased with 44 % while the capital costs are reduced with 23 %. These results confirm the statement that reducing the need for heat transfer is a main objective of the design of high temperature fuel cell systems. This understanding should be guiding for fuel cell development. The equivalent cell resistance is another parameter that has been investigated in the considered study. A reduction of the equivalent cell resistance increases the electrical output of the cell and decreases the generated heat and therefore the power from the bottoming steam cycle. Improving fuel cell performance in a system with bottoming cycle has a limited effect on overall system efficiency as the power increase of the fuel cell stacks is partly compensated by the reduced power from the bottoming cycle. For the configuration shown in figure 13 the exergy loss due to heat transfer is rather high and therefore reducing the cell



## Figure 14. Cell voltage and net system efficiency versus the equivalent cell resistance

resistance has still a serious effect on overall system efficiency, as shown in figure 14. Reducing the equivalent cell resistance from 1.0 to 0.5  $\Omega$ cm<sup>2</sup> increases the cell voltage from 0.59 to 0.71 V and overall system efficiency from 62 till 69 %. However increasing the exergy efficiency of the heat transfer system will limit the effect of reduced cell resistance.

Combining SOFC's with gas turbines for intermediate to large scale power plants seems to be a very attractive option for the future application of high temperature fuel cells. Several system studies are devoted to this subject. In [21] it was concluded that such a system can achieve electrical efficiencies of approximately 75 % (LHV) when applying conventional gas turbine technology. The major part of the electricity (76 %) is generated by the fuel cell stacks. It is expected that on the long term electrical efficiencies up to 80 % will become possible due to further developments of gas turbine and SOFC technologies as well as careful optimisation of the system configurations. In the combined SOFC-gas turbine system the SOFC replaces actually the combustion chamber of the gas turbine, thus reducing the large exergy losses of combustion. The outlet gasses of the SOFC have temperatures corresponding with the operating temperature of the SOFC (800 -1000 °C). The gas temperature can be increased further by burning the residual fuel from the anode in an afterburner with the cathode off-gas as the oxidiser. This will result in appropriate temperature conditions for modern gas turbines. The SOFC as well as the combined SOFC-gas turbine system can also be seen as an efficient heat generator for industrial processes. Whether the SOFC or the SOFC-gas turbine is preferred will also depend on the temperature level of the heat. Bottoming cycles, steam cycles of organic rankine cycles, can be included to adapt the plant design more carefully to the requested temperature levels of the demanded heat.

#### SUMMARY AND CONCLUSIONS

Thermal power plants have large exergy losses caused by the thermal conversion of fuel. These losses are inherent to the conversion process. Elimination or reduction of these losses will require conversion processes with seriously lower irreversibility's. Electrochemical conversion therefore provides an opportunity for a substantial reduction of thermodynamic losses in case of generation of useful energy like power and heat.

The development history of fuel cells has shown that accompanying system studies are necessary to define boundary conditions this new technology has to fulfil. Exergy analysis can provide true insight into the thermodynamic losses of the system and can be helpful in searching system improvements. System evaluation software with high flexibility to system design and plenty facilities for analysing the thermodynamic performance of energy conversion systems, like facilities for exergy analysis, can be very useful.

The performance of low temperature fuel cell systems, like PEMFC systems, using natural as the primary fuel is mainly depending on the losses in the cell stack and in fuel processing. During the last decades large progress has been made in the development PEM fuel cells. The development of high temperature PEMFC's might affect the requirements to the preceding fuel processing plant, but electrochemical conversion will enable a reduction of losses due to fuel processing anyhow. PEMFC's are supposed to be suitable also for micro-CHP application. However micro-CHP has to compete with electrical driven heat pumps; increased efficiencies of electricity generation will favour the position of heat pumps and will make the use of micro-CHP questionable.

The performance of high temperature fuel cell system is strongly determined by exergy losses due to heat transfer. Therefore the following general guidelines have to be considered during the design of HT fuel cell systems:

- 1. reduce the generation of heat in the fuel cell (minimise internal resistance, apply multistage oxidation, etc.),
- 2. use heat directly (for internal reforming or in integrated gas turbine),
- 3. avoid heat transfer in heat exchangers (by applying cathode and anode recycle),
- 4. enable heat exchange with sufficiently small temperature differences (optimise heat integration).

The application of high temperature fuel cells will require an optimum integration of electrochemical and thermal power conversion technologies to obtain power plants with considerable higher efficiencies. Actually high temperature fuel cells should replace the combustion process in systems for the generation of power and heat, thus reducing the large thermodynamic losses that are inherent to thermal fuel conversion. Integration with suitable gas turbine processes and bottoming cycles will enable optimum adaptation to the temperature level of the demanded heat. The introduction of fuel cells is considered to be a first step in the integration of electrochemical conversion in future energy conversion systems.

High temperature fuel cells will be necessary to obtain conversion efficiencies up to 80 % in case of large scale electricity production in the future. The fast growing energy consumption makes the need for such high conversion efficiencies apparent.

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