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TOWARDS MULTI FUEL SOFC PLANTS

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ABSTRACT

Complete Solid Oxide Fuel Cell (SOFC) plants fed by several different fuels are suggested and analyzed. The plants sizes are about 10 kW which is suitable for single family house with needs for both electricity and heat. Alternative fuels such as, methanol, DME (Di-Methyl Ether) and ethanol are also considered and the results will be compared with the base plant fed by Natural Gas (NG). A single plant design will be suggested that can be fed with methanol, DME and ethanol whenever these fuels are available. It will be shown that the plant fed by ethanol will have slightly higher electrical efficiency compared with other fuels. A methanator will be suggested to be included into the plants design in order to produce methane from the fuel before entering the anode side of the SOFC stacks. Increasing methane content will decrease the needed compressor effect and thereby increase the plant power.
Keywords: SOFC, multi fuel, methanator, methanol, DME, ethanol

INTRODUCTION

The Solid Oxide Fuel Cell (SOFC) is an electro-chemical reactor currently under development by several companies for power-heat generation application. Depending on the type of the electrolyte they are operating at temperature levels of more than about 750°C up to 1000°C. Due to material cost, the lower temperature alternative is now being developed for market entry during this decade. This would also be in advantage for the BoP (Balance of Plants) components.

The biggest advantage of the SOFC in comparison with other types of fuel cells may be in its flexibility in using different types of fuels. However, in planar SOFCs one needs to pre-process most kind of fuels in order to break down the heavier hydro-carbons which may otherwise poison the solid oxide fuel cells. The sulphur content in the fuels must also be removed before entering the anode side of the SOFC. Such pre-processing can be done in two different catalytic reactors operating at different temperature levels indicated by reactor manufacturers.

SOFC – based power plants have been studied for a while and some companies, such as Wärtsilä, are trying to realize such systems for CHP (Combined Heat and Power) applications; see e.g. [1]. The SOFC is also combined with CC (Combined Cycles) in the literature to achieve ultra high electrical efficiencies, see e.g. [2–3]. Due to the current operating temperature of the SOFC stacks, hybrid SOFC and GT (Gas Turbine) systems have also been studied extensively in the literature, e.g. in [4] for CHP (Combined Heat and Power). Characterization, quantification and optimization of hybrid SOFC–GT systems have been studied by e.g. [5]. In [6] modeling results are compared with measured data for a 220 kW hybrid planar SOFC–GT power plant. Details on design, dynamics, control and startup of such hybrid power plants are studied in [7]. While hybrid SOFC–GT plants have been extensively studied by many researchers, the investigations on combined SOFC and ST (Steam Turbine) are very limited see [8].

Fuel pre-reforming can be done in different reactors such as ASR (Adiabatic Steam Reformer) and CPO (Catalytic Partial Oxidation). The disadvantages of an ASR reactor is that it needs superheated steam during start–up (depending on the operating temperature of the reactor, i.e. 400°C), which is an extremely power consuming

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procedure. During normal operation steam is available after the anode side of the SOFC stacks, which can be recycled into the system. In a CPO reactor some of the fuel is burned to reach the reformer temperature (exothermic), which in turn decreases the plant efficiency. In this study both ASR and CPO reforming processes are applied for the plant fed by natural gas, which is assumed to be the base cases when comparison with alternative fuels is carried out.

SOFC plants fed by alternative fuels are designed and suggested. The alternative fuels are assumed to be methanol, DME and ethanol which do not produce any CO_2 at consumption side. A methanator is applied in these plants to increase the amount of methane prior to SOFC anode side. Increasing the amount of methane increases the extent of the endothermic reforming reactions and thus decreases the excess air flow which is used to cool down the SOFC stacks.

METHODOLOGY

The results of this paper are obtained using the simulation tool DNA (Dynamic Network Analysis), see [9], which is a simulation tool for energy system analysis. In DNA the mathematical equations include mass and energy conservation for all components, as well as relations for thermodynamic properties of the fluids involved. The program is written in FORTRAN. The component library includes models of various components such as; heat exchangers, burners, turbo machinery, dryers and decanters, energy storages engines, valves, controllers, as well as more specialized components and utility components. The user may also implement additional components. For example fuel cell model, methanator model and reformer model are included in this study as is going to be described below.

Modeling of SOFC

The SOFC model used in this investigation is based on the planar type developed by DTU-Risø and TOPSØE Fuel Cell. The model is calibrated against experimental data in the range of 650°C to 800°C (operational temperature) as described in

[10]. The model matches exactly the experimental data. The outlet temperatures (anode and cathode sides) are assumed to be the same as the operating temperature. The operational voltage (E_{FC}) is expressed as

$$E_{FC} = E_{Nernst} - \Delta E_{act} - \Delta E_{ohm} - \Delta E_{conc} - \Delta E_{offset} \tag{1}$$

where E_{Nernst}, ΔE_{act}, ΔE_{ohm}, ΔE_{conc}, ΔE_{offset} are the Nernst ideal reversible voltage, activation polarization, ohmic polarization, concentration polarization and the offset polarization respectively. The activation polarization is expressed as Butler–Volmer equation (see [11]). The activation polarization is isolated from other polarization to determine the charge transfer coefficients as well as exchange current density from the experiment by curve fitting technique. It follows,

$$\Delta E_{act} = \frac{RT}{(0.001698T - 1.254)F} x$$
$$\sinh^{-1}\left[\frac{i_d}{2(13.087T - 1.096 \times 10^4)}\right] \tag{2}$$

where R, T, F and i_d are the universal gas constant, operating temperature, Faradays constant and current density respectively. Ohmic polarization depends on the electrical conductivity of the electrodes as well as the ionic conductivity of the electrolyte and can be described as

$$\Delta E_{ohm} = \left(\frac{t_{an}}{\sigma_{an}} + \frac{t_{el}}{\sigma_{el}} + \frac{t_{ca}}{\sigma_{ca}}\right)i_d \tag{3}$$

where t_{an} = 600 μm, t_{el} = 50 μm and t_{ca} =10 μm are the anode thickness, electrolyte thickness and cathode thickness respectively. σ_{an}, σ_{el} and σ_{ca} are the conductivity of anode, electrolyte and cathode respectively.

$$\sigma_{an} = 10^5 \tag{4}$$

$$\sigma_{ca} = \frac{5.760 \times 10^7}{T} \exp\left(-\frac{0.117}{8.617 \times 10^{-5}T}\right) \tag{5}$$

$$\sigma_{el} = 8.588 \times 10^{-8}T^3 - 1.101 \times 10^{-4}T^2 + 0.04679T - 6.54 \tag{6}$$

Concentration polarization is dominant at high current densities for anode – supported SOFC, wherein insufficient amounts of reactants will be transported to the electrodes and the voltage will

then reduce significantly. Neglecting the cathode contribution (see e.g. [12]), it can be modeled as

$$\Delta E_{conc} = B\left(\ln\left(1 + \frac{p_{H2}i_d}{p_{H2O}i_{as}}\right) - \ln\left(1 - \frac{i_d}{i_{as}}\right)\right) \quad (7)$$

where B is the diffusion coefficient which is calibrated against experimental data and found to be,

$$B = \left(0.008039X_{H2}^{-1} - 0.007272\right)\frac{T}{T_{ref}} \quad (8)$$

In the above equations p_{H2} and p_{H2O} are the partial pressures for the H_2 and H_2O respectively, while T_{ref} is the reference temperature (1023 K). The anode limiting current density is defined as

$$i_{as} = \frac{2F\,p_{H2}D_{bin}V_{an}}{RT\,t_{an}\tau_{an}} \quad (9)$$

where V_{an} and τ_{an} are the porosity and tortuosity of the anode and are the physical characteristics as 30% and 2.5 μm in the experimental setup. The binary diffusion coefficient is given by

$$D_{bin} = \left(-4.107\text{x}10^{-5}X_{H2} + 8.704\text{x}10^{-5}\right)\left(\frac{T}{T_{ref}}\right)^{1.75}\frac{P_{ref}}{P} \quad (10)$$

which is also calibrated against the experimental data. P_{ref} is the reference pressure as 1.013 bar and X_{H2} is the mass reaction rate of H_2. Finally the current density i_d is directly proportional to the amount of reacting hydrogen according to the Faraday's law;

$$i_d = \frac{\dot{n}_{H2}\,2F}{A} \quad (11)$$

where $\dot{n}_{H2}$ is molar reaction rate of H_2. The area A is the physical property of the cell and is 144 cm^2.

Modeling of Methantor

The reforming process is assumed to reach chemical equilibrium by minimizing the Gibbs free energy as described in [14]. A similar procedure is also applied for modeling the methanator. The Gibbs free energy of a gas (assumed to be a mixture of k perfect gases) is given by

$$\dot{G} = \sum_{i=1}^{k}\dot{n}_i\left[g_i^0 + RT\ln(y_i p)\right] \quad (12)$$

where g^0, R, T and y_i are the specific Gibbs free energy, universal gas constant, gas temperature and molar fraction respectively. Each atomic element in the inlet gas is in balance with the outlet gas composition, which yields the flow of each atom has to be conserved. For N elements this is expressed as

$$\sum_{i=1}^{k}\dot{n}_{i,in}\,\mathbf{A}_{ij} = \sum_{m=1}^{w}\dot{n}_{m,out}\,\mathbf{A}_{mj} \quad \text{for } j = 1,N \quad (13)$$

The N elements correspond to H, C and O in this pre-reforming process. $\mathbf{A}_{mj}$ is the number of atoms of element j (H, C, O, N) in each molecule of entering compound i (H_2, CH_4, CO, CO_2, H_2O, O_2, N_2 and Ar), while $\mathbf{A}_{ij}$ is the number of atoms of element j in each molecule of leaving compound m (H_2, CH_4, CO, CO_2, H_2O, N_2 and Ar). The minimization of Gibbs free energy can be formulated by introducing a Lagrange multiplier, μ, for each of the N constraints obtained in Eq. (13). After adding the constraints, the expression to be minimized is then

$$\phi = \dot{G}_{tot,out} + \sum_{j=1}^{N}\mu_j \times \left(\sum_{i=1}^{k}\dot{n}_{i,out}A_{ij} - \sum_{m=1}^{w}\dot{n}_{m,in}A_{mj}\right) \quad (14)$$

The partial derivation of this equation with respect to $\dot{n}_{i,out}$ can be writes as

$$\frac{\partial\phi}{\partial\dot{n}_{i,out}} = g_{i,out}^0 + RT\ln(y_{i,out}p_{out}) + \sum_{j=1}^{k}\mu_j\mathbf{A}_{ij} \quad \text{for } i = 1,k \quad (15)$$

At the minimum each of these is then zero. An additional equation is added to make sure that the summation of molar fractions of the outlet gases is to be the unity.

RESULTS

Plants Fed by Natural Gas

The first configuration studied is shown in Fig. 1a. The fuel (NG) is preheated in a heat exchanger before it is sent to a desulphurization unit to remove the sulfur content in the NG. This unit is assumed to be a catalyst, operating at a temperature of 200°C. Thereafter the heavier carbon contents in the NG are broken down in a CPO type pre-reformer catalyst. Before that the fuel must be preheated again to reach to the operational temperature of the CPO catalyst. The CPO catalyst needs additional air which is supplied by a small pump as shown in the figure. This air is also preheated before entering the pre-reformer. It is assumed that the supplied NG is pressurized and therefore no pump is needed for the fuel. The pre-reformed fuel is now sent to the anode side of the SOFC stacks. Due to the exothermal nature of the CPO catalyst, no preheating of the fuel is required after the reformer. The fuel has a temperature of about 650°C before entering the stacks. The operating temperature of the SOFC stacks as well as outlet temperatures is assumed to be 780°C. The used fuel after the SOFC stacks is used to preheat the fuel as well as the air needed for CPO reformer by using two separate heat exchangers.
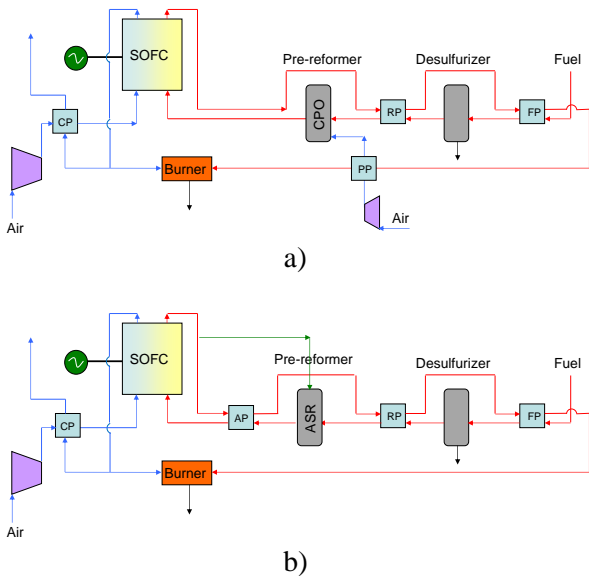

a)

b)

Figure 1. The complete SOFC plant fed by natural gas, a) with CPO reformer b) with ASR reformer.

On the other side, air is compressed in a compressor and then preheated in a recuperator to about 600°C before entering the cathode side of the SOFC stacks. Part of the air after the cathode side is used to preheat the incoming air. Thus it is assumed that the entering temperatures shall not be less than 600°C. Lower entering temperature may shut down the stacks automatically. Since the fuel in the SOFC stacks will not be utilized completely the rest of the fuel together with the air coming out of the cathode side are sent to a burner (catalytic burner) for further burning.

As mentioned earlier the ASR reformer needs super heated steam for operation. Such steam must be supplied to the reformer externally during start–up. However, during normal operation steam is available after SOFC stacks due to reactions of hydrogen and oxygen. Therefore, the stream after the anode side of SOFC is recycled as shown in Fig. 1b. There are three alternatives for such a recirculation unit, a pump, a turbocharger and an ejector. In a real plant, due to high temperature of this stream (more than 700°C) the cost of a pump working will be rather expensive. This is also true for a turbocharger which is working at such mass flows and pressures. Moreover, using an ejector brings up problems associated with the size and dimensioning of the ejector (due to combination of pressure drop and mass flows). Based on these facts and for the sake of simplicity an ejector is used in this investigation. In order to have a proper oxygen-carbon-ratio (~2), 50% of the anode outlet is recycled. Due to endothermic nature of the ASR reformer a heat exchanger is used to raise the temperature of the reformed fuel to 650°C prior to anode side of the SOFC stacks.

The main parameters for the plant are set in table 1. Number of stacks is assumed to be 10 meaning that each stack would produce 1kW electric power with 74 cells. The aim is to produce 10kW net power which is enough for a family house in Scandinavian countries. Thus electric power production by SOFC stacks is set to 10kW.

The pressure drops in the cathode sides as well as the component on the path of air (heat exchanger), is assumed to be 0.005 bars. The pressure drop in the anode side as well as the components in the path of fuel (heat exchangers, desulfurizer and reformer) is assumed to be 0.001 bars. These

values are the setting values for the program, however, pressure drops are a function of channel sizes and mass flows and the channel geometry is not kwon. Therefore, these values are calculated based on the available data for each channel mass flow and dimensions.

The SOFC plant provides direct current and must be converted to AC through a converter. Further, the efficiency of the DC/AC converter is assumed to be 100%. In reality there would be some losses trough the converter and efficiencies of 97% could be assumed for plants of such sizes studied in this investigation. The inlet temperatures of both pre-reformers are assumed to be 400°C. For the CPO case the inlet air to the pre-reformer is thus preheated to this temperature as well.

Set points Parameter	CPO	ASR
Compressor intake T [°C]	25	25
Compressor $\eta_{isentropic}$	0.60	0.60
Compressor $\eta_{mechanical}$	0.95	0.95
SOFC cathode inlet T [°C]	600	600
SOFC cathode outlet T [°C]	780	780
SOFC utilization factor	0.80	0.80
SOFC number of cells	74	74
SOFC number of stacks	10	10
Δp in the fuel side [bar]	0.005	0.005
Δp in the air [bar]	0.001	0.001
Fuel inlet temperature [°C]	25	25
Desulfurizer operation T [°C]	200	200
SOFC anode inlet T [°C]	650	650
SOFC anode outlet T [°C]	780	780
Burner efficiency	0.97	0.97

Table 1. Main parameters for design point calculations.

The main calculated parameters are provided in Table 2.

Parameter	CPO	ASR
Net power output [kW]	9.85	9.85
SOFC plant efficiency [%]	48.1	55.8
Fuel consumption [kJ/s]	20.48	17.66
Net power consumption [W]	147.2	152.4
Burner outlet T [°C]	1170.0	951.7

Table 2. Net powers and efficiencies for the plants fed by natural gas.

The plants net powers and thermal efficiencies (based on LHV, Lower Heating Value) are shown in Table 2. The net power output of both plants is similar. The efficiency of the ASR plant is considerably higher than the corresponding CPO plant. The reason is the additional air needed for CPO reformer which is provided by the reformer compressor. This is reflected in higher fuel consumption for the CPO plant. Further, the temperature of the burner for the CPO plant is considerably higher than the ASR plant. This means that the heat generation for the CPO would be more than the ASR plant. This of course is justified by lower plant efficiency.

As mentioned previously, the inlet temperature of both reformers are assumed to be 400°C. However, one could assume other temperatures than this; therefore, Fig 2 is presented to study the plant efficiency versus reformer inlet temperature. As can be seen the plant efficiency with ASR pre-reformer does not change significantly when the inlet temperature is increased to 550°C. On the other hand, the plant efficiency increases when the CPO pre-reformer inlet temperature is increased. As mentioned previously, the reason that the inlet reformers temperatures assumed to be 400°C was that the oxygen-carbon-ratio for the CPO and steam-carbon-ratio for ASR would be about 2, thus carbon deposition is avoided. Note that, increasing CPO inlet temperature means that supplied air to the reformer is decreased, while recycle for ASR kept constant.
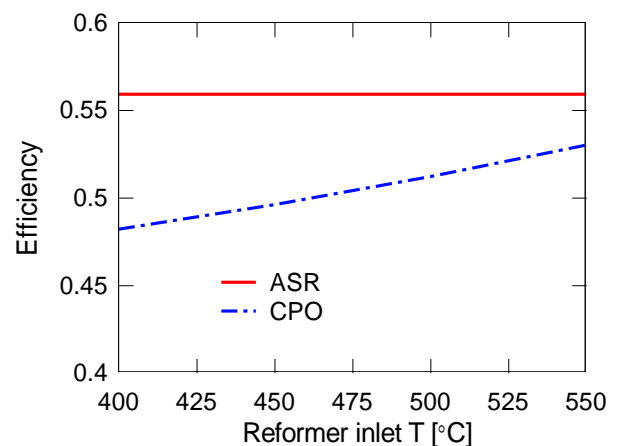

Figure 2. Plant efficiency as function of reformer inlet temperature.

Plants Fed by Alternative Fuels

The alternative fuels to be used here are methanol, ethanol and DME. These fuels can be produced from different sources such as biomass, natural gas and coal. At production side there will be CO_2 emission wherein its amount depends on the production source. In this way emission is centralized which means that the CO_2 emission can be captured. However, such capturing utility at consumer side will be difficult and extremely costly. From now on the phrase alternative fuels refer to methanol, DME and ethanol. Fuel properties are shown in Table 3.

Fuel	Chemical composition	LHV [MJ/kg]	LHV [MJ/L]
Methanol	CH3OH	19.9	15.8
DME	CH3OCH3	28.8	19.3*
Ethanol	C2H5OH	28.9	22.8

Table 3. Comparison between the fuels.
*Liquid (above 5 bar).

All these fuels can be directly fed to the anode side of the fuel cell without the need for desulfurizer and pre-reformer, since there is no sulfur and heavier hydrocarbons in these fuels. Thus the complete plant would be much simpler than the corresponding plant fed by natural gas, see Fig. 3. Both the fuel and air are preheated to 600°C before entering the SOFC stacks. In order to increase the oxygen-carbon-ratio some of the fuel is recycled after the anode side of the SOFC stacks.
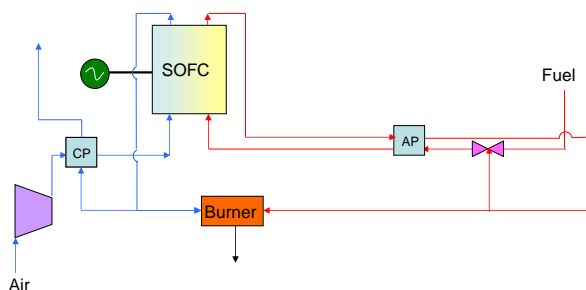

Figure 3. The complete SOFC plant fed by alternative fuels, methanol, DME and ethanol.

In order to have a fair comparison, number of stacks is chosen to be the same as in the case for natural gas (10 stacks). Other assumptions are also the same as in the case for natural gas as shown in Table 1, such as pressure drops in the heat exchangers and burner efficiency.

Power production, plant efficiency and amount of fuel recycle (after anode) are presented in Table 4. As can be seen from table 3., the power produced by DME and ethanol are almost similar and slightly higher than the case with natural gas, while the power produced by methanol is slightly lower than the case with natural gas.

Fuel	Efficiency [%]	Net Power [kW]	Recycle [%]
Methanol	49.3	9.73	41
DME	50.3	9.94	37
Ethanol	54.1	9.93	38

Table 4. Comparison between the fuels.

The plant fed by ethanol has the best efficiency (~54%) while the plant fed by methanol has the lowest efficiency (~49%).

It might be interesting to mention that the power production by ethanol and DME are rather similar but the efficiency of the plant fed by ethanol is much higher than the case for DME. The reason is that the chemical reactions in the SOFC stacks are exothermic meaning that heat is going to be produced. Therefore, in order to keep the temperature of the SOFC stacks at the same level additional excess airflow is needed to cool down the SOFC stacks. This excess air is provided by the air compressor. For the case fed by DME it requires much higher air flow than for the case fueled by ethanol to cool down the SOFC stacks.

Effect of Methanator

As discussed above the mass flow of the air is considerably higher than the stoichiometry value and the reason is to cool down SOFC stacks and keep their temperature at the desired value. It might then be interesting to discus possibilities to decrease the air mass flow somehow without affecting the operating temperature of the fuel cells. One possibility is to increase the amount of methane in the fuel prior to the anode side. Due to reforming reactions (methane and water, CO_2) and its endothermic (demanding heat) property then

heat can be absorbed from the cells. Increasing methane content in the fuel will then result in more such reaction, which in turn results in more heat will be absorbed. Thus the air flow will be decreased. This can be achieved by adding an adiabatic Methanator into the plant as shown in Fig. 4. Fuel is preheated to 300°C and then is entered into the Methanator wherein the amount of methane is increased before entering to the anode side of the SOFC stacks. To avoid carbon deposition additional steam (available after SOFC stacks) is recycled and then mixed with incoming fuel as shown in the figure.
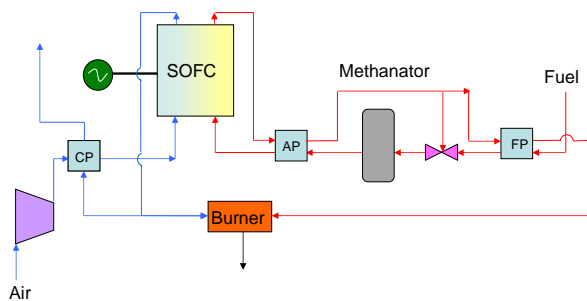

Figure 4. Suggested complete SOFC plant with Methanator when alternative fuels are used.

For the sake of safety a heat exchanger is used after the Methanator in case that the temperature of the fuel is not less than about 600 ~ 650°C. In fact, the temperature after the Methanator is about 600°C, which alleviates the need for the anode pre-heater (AP).
The results for such plant with the adiabatic Methanator is shown in Table 4 in terms of plant efficiency, net power produced and fuel consumption. As can be seen the plant efficiency fed by ethanol does not change considerably whether a Methanator is used or not. However, the plant efficiency fed by DME is increased slightly (by 1 point percentage) when a Methanator is used. Finally, the plant fed by methanol increased by about 2 point percentage when a Methanator is used compared with the corresponding plant without Methanator. Thus it can be concluded that the use of Methanator has a significant effect on plant efficiency and depends on the fuel type and molecular bounds.
Table 5 shows also that the plant fed by methanol needs significantly higher fuel mass flow (about

31% more) than DME and ethanol to provide the same net power of about 10kW.

Fuel	Efficiency [%]	Net Power [kW]	Fuel [kg/h]
Methanol	51.3	10.13	3.5712
DME	51.3	10.13	2.466
Ethanol	54.2	10.14	2.466

Table 5. Comparison between the fuels in a plant with Methanator.

CONCLUSIONS

A SOFC plant with about 10kW net power running by several fuels is studied. In the plant fed by natural gas both ASR and CPO pre-reformers were used to quantify their influence on plant efficiency. It was shown that the plant with ASR reformer had higher efficiency than the plant with CPO pre-reformer, 55.8% versus 48.1%. The reason was that the CPO pre-reformer burned some of the fuel to reach the reactor temperature and therefore, less fuel was available for the SOFC stacks and consequently less power was produced by the plant compared to corresponding ASR plant.
A general plant fed by alternative fuels (methanol, DME and ethanol) was also studied to compare the plants in terms of plant efficiency. It was shown that ethanol had a significantly higher plant efficiency compared with the corresponding plants fed by methanol and DME, 54.1% compared to 49.3% and 50.3%, respectively.
It was also concluded that using an adiabatic Methanator could increase plant efficiency fed by methanol significantly, while its effect on plant efficiency fed by DME was marginal. The use of adiabatic Methanator had no significant effect on plant efficiency when the plant was fed by ethanol. Such conclusions are valid under assumptions made here, such as number of stacks, temperature after the methanator or amount of recycled fuel. Other assumptions may slightly change the conclusions drawn here which are not discussed here.

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