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SOFC-MGT hybrid power plants fuelled by methanol and DME

Daniele Cocco · Vittorio Tola

Abstract This work analyses the performance of hybrid power plants based on solid oxide fuel cells integrated with micro-gas turbines (SOFC-MGT). Internally and externally reformed SOFC-MGT systems fuelled with methane, methanol, ethanol and DME have been compared. The results show that simply replacing methane with methanol, ethanol or DME in internally reformed SOFC-MGT systems slightly reduces efficiency and power output. In contrast, using methanol and DME in externally reformed SOFC-MGT hybrid plants can lead to efficiency improvement with respect to internally reformed hybrid plants fuelled by methane, especially for the higher values of the fuel utilization factor (higher than about 70% for methanol and 80% for DME). Finally, the main operating parameters of the fuel reforming section (temperature and steam-to-carbon ratio, SCR) must be carefully chosen in order to optimise the hybrid plant performance.

Keywords DME reforming · External reforming · Ethanol reforming · Methane reforming · Methanol reforming · Micro-gas turbines · Solid oxide fuel cell (SOFC)

1 Introduction

Fuel Cells (FC) and Micro-Gas Turbines (MGT) are one of the most interesting options for distributed power generation, due to their high efficiency and low pollutant emissions. Moreover, high temperature fuel cells (Molten Carbonate Fuel Cell, MCFC and Solid Oxide Fuel Cells, SOFC) and MGT can be suitably integrated to improve conversion efficiency. In particular, as the turbine inlet temperature of MGT power plants (about 900–1,000 °C) is very similar to the gas exit temperature of SOFC stacks, SOFC-MGT hybrid power plants can achieve efficiency values higher than 60% even for small power outputs (200–500 kW) [1–9].

Hybrid SOFC-MGT power plants currently developed are fuelled by natural gas, mainly composed by methane, and the heat required by the endothermic methane reforming reactions is supplied directly by the fuel cell stack itself at about 800–900 °C. On the other hand, some alternative fuels (methanol, ethanol, di-methyl-ether, etc.) are under evaluation for distributed power generation [10–14]. These fuels are useful energy carriers rather than primary energy sources and they can be produced from a wide range of primary fuels (natural gas, coal, biomass, etc.) through gasification or reforming plants integrated with chemical synthesis processes. Ethanol can also be produced from sugar crops (sugar cane, sugar beet, etc.) or starch crops (corn, wheat, potatoes, etc.) by fermentation. These alternative fuels are liquid at ambient temperature (even though DME needs to be liquefied under pressure at ambient temperature) so they can be readily handled, stored and transported. Moreover, methanol, ethanol and DME are free from sulphur, heavy metals, and other impurities so they can be used in fuel cells, gas turbines and internal combustion engines, with high conversion efficiency and low pollutant emissions. In the last few years many projects on methanol and DME production have been announced in countries like Japan, Korea, Taiwan, India, China, etc. Moreover, the world's ethanol production from sugar cane and corn is rapidly increasing (40 billion litres in 2004, almost double the 2000 production).

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Methanol, ethanol and DME can be directly used in internally reformed SOFC-MGT power plants, even though simply replacing methane with these fuels slightly reduces efficiency and power output. However, the low reforming temperature of methanol and DME (200–250 °C for methanol and 250–300 °C for DME) allows siting at the reformer outside the stack, thus improving the low temperature exhaust heat recovery. In contrast, methane and ethanol are unsuitable for externally reformed SOFC stacks, as their reforming temperature ranges from 700 to 900 °C.

Starting from the results of earlier studies concerned with the use of methanol and DME in SOFC-MGT hybrid power plants [15–17], in this paper the utilization of methane, methanol, ethanol and DME in SOFC-MGT hybrid power plants with internal reforming and external reforming has been analysed.

2 SOFC-MGT hybrid systems modelling and assumptions

The performance of the hybrid SOFC-MGT power plants has been evaluated using ASPEN PLUS^{TM} simulation software, version 12.1 [18]. In particular, the ASPEN model library, which includes many standard components used by energy conversion systems (heat exchangers, pumps, turbines, reactors, etc.), has been integrated with a dedicated fuel cell model, developed by the authors [15–17].

Figure 1 shows the configuration considered for the internally reformed SOFC-MGT hybrid power plant. This configuration is similar to recently proposed hybrid SOFC-MGT plants and is based on an internally reformed SOFC stack integrated with a recuperated micro-gas turbine [1–4]. The air is first pressurized by the compressor (C), and then heated in the recuperator (RC) before being supplied to the SOFC stack. After expansion through the turbine (T), the main portion of the SOFC exhaust gas is cooled in the recuperator, whereas the remainder is cooled in the fuel heat exchanger (FH) for pre-heating the fuel. When the SOFC-MGT plant is fuelled by liquid fuels (methanol, ethanol or DME), the heat from the two flue gas streams exiting the RC and FH is used in the fuel vaporizer (FV), with a further low temperature waste heat recovery. Therefore, the SOFC-MGT power plant comprises the fuel vaporizer (dotted lines of Fig. 1) only when methanol, ethanol or DME are used as primary fuels. In the latter case, these fuels enter the FH at their vaporization temperature, whereas methane enters the FH at ambient temperature.

As known, methanol reforming is almost complete (that is fuel conversion is higher than about 99%) above 200–
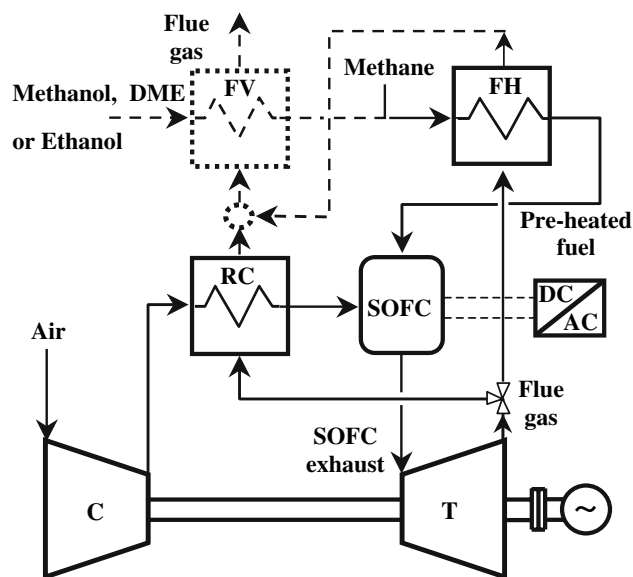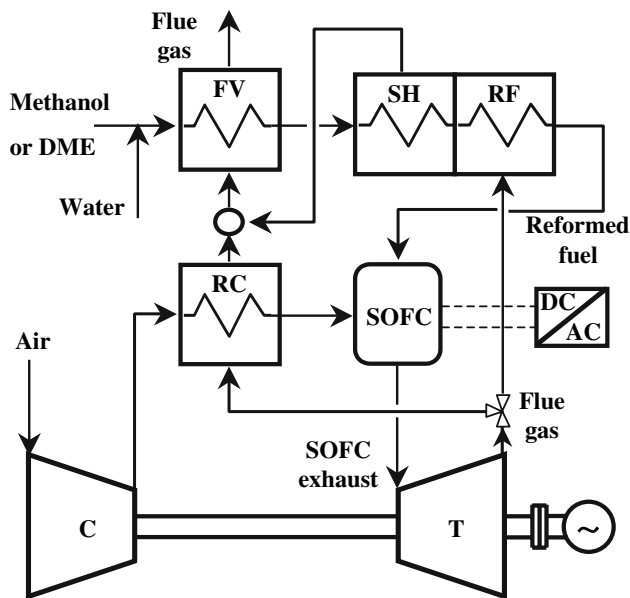

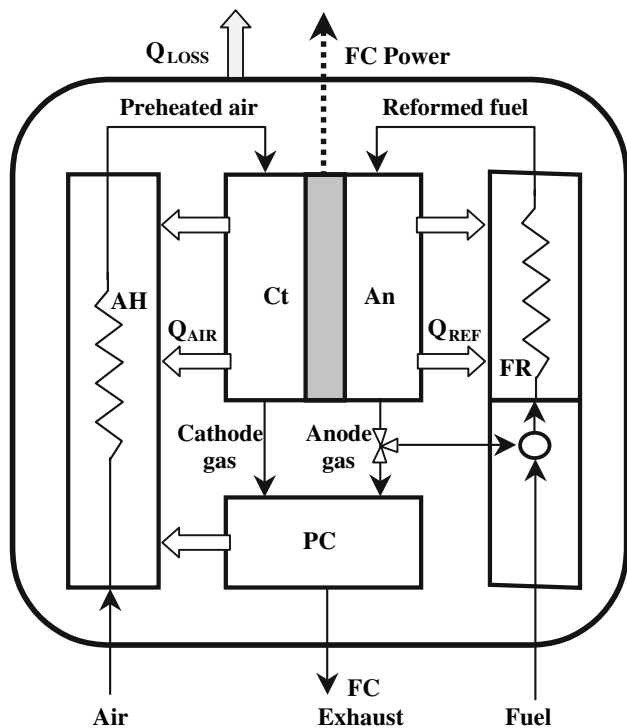**Fig. 1** Configuration of the SOFC-MGT hybrid power plant with internal reforming

250 °C and DME reforming above 250–300 °C. In contrast, the complete reforming of methane and ethanol requires higher temperatures 700–900 °C [19–26]. The lower reforming temperature of methanol and DME allows siting at the reformer outside the stack, thus improving low temperature exhaust heat recovery, as the RC exit gas is available at about 250–300 °C. For this reason, methanol and DME are suitable fuels for externally reformed SOFC-MGT hybrid systems.

Figure 2 shows the configuration considered for the SOFC-MGT hybrid power plant with external reforming. In this case, the fuel vaporizer is fed by the fuel/water mixture, and the FH heat exchanger of Fig. 1 has been replaced by the external reforming section (RF + SH). The latter comprises the catalytic steam reforming section (RF), where the endothermic steam reforming reactions take place, and the super-heating section (SH), where the vaporized water/fuel mixture is heated up to the required reforming temperature.
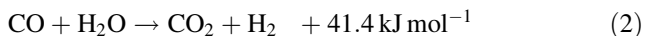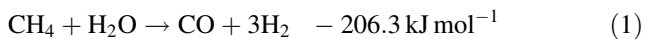
Figure 3 shows the scheme of the internally reformed SOFC stack. The fuel (methane, methanol, ethanol or DME) is mixed with part of the anode exhaust gas (containing water produced by hydrogen oxidation) to provide the Steam-to-Carbon Ratio (SCR) required by the subsequent reforming process. The mixture flows through the internal fuel reformer (FR), where the endothermic fuel reforming reactions take place. The reformed fuel gas composition has been evaluated under the assumption of thermodynamic equilibrium, calculated at the stack operating temperature (here assumed equal to 900 °C). In particular, for methane the following steam reforming reactions have been considered [1–2, 14]:
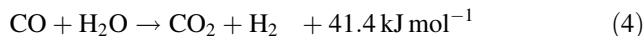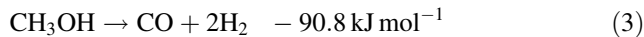
Fig. 2 Configuration of the SOFC-MGT hybrid power plant with external reforming
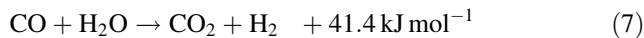

Fig. 3 Conceptual scheme of the SOFC stack

$$CH_4 + H_2O \rightarrow CO + 3H_2 \quad -206.3\,kJ\,mol^{-1} \quad (1)$$
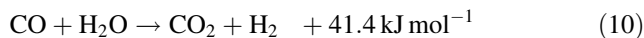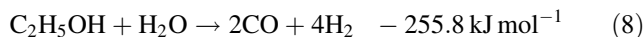
$$CO + H_2O \rightarrow CO_2 + H_2 \quad +41.4\,kJ\,mol^{-1} \quad (2)$$

The methanol reforming process has been described by means of the following reactions [19–20]:

$$CH_3OH \rightarrow CO + 2H_2 \quad -90.8\,kJ\,mol^{-1} \quad (3)$$

$$CO + H_2O \rightarrow CO_2 + H_2 \quad +41.4\,kJ\,mol^{-1} \quad (4)$$

The DME reforming process has been described by means of the following reactions [21–22]:

$$CH_3OCH_3 + H_2O \rightarrow 2CH_3OH \quad -22.2\,kJ\,mol^{-1} \quad (5)$$

$$CH_3OH \rightarrow CO + 2H_2 \quad -90.8\,kJ\,mol^{-1} \quad (6)$$

$$CO + H_2O \rightarrow CO_2 + H_2 \quad +41.4\,kJ\,mol^{-1} \quad (7)$$
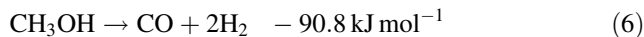
The ethanol reforming process can be described by means of the following reactions [23–26]:

$$C_2H_5OH + H_2O \rightarrow 2CO + 4H_2 \quad -255.8\,kJ\,mol^{-1} \quad (8)$$

$$CO + 3H_2 \rightarrow CH_4 + H_2O \quad +206.3\,kJ\,mol^{-1} \quad (9)$$

$$CO + H_2O \rightarrow CO_2 + H_2 \quad +41.4\,kJ\,mol^{-1} \quad (10)$$

According to the above reforming reactions, the complete conversion of 1 mole of methane requires at least 2 moles of water. Similarly, the complete conversion of 1 mole of methanol requires at least 1 mole of water, while the complete conversion of each mole of DME and ethanol requires at least 3 moles of water. The corresponding stoichiometric SCR is 2 for methane, 1 for methanol and 1.5 for ethanol and DME. Too small SCRs are to be avoided as they can lead to carbon formation, whereas too high SCRs lead to H_2 dilution [14, 19].

Obviously, the fuel reformer (FR) and the anode gas recirculation are not required for externally reformed hybrid systems, as the reformed fuel gas is directly produced by the external reforming section (RF + SH). In this case, the steam reforming process has been conventionally carried out in two stages. In the first stage (SH) the vaporized water/fuel mixture produced by the FV heat exchanger is superheated to the desired reforming temperature T_{RF} without considering any chemical reaction. In the second stage (RF) the mixture reforms at constant temperature. The reformed gas composition has been evaluated under the assumption of thermodynamic equilibrium and the equilibrium constants for reactions (3–7) have been calculated by assuming the equilibrium temperature T_{EQ}. In particular, at high reforming temperatures (over 250 °C for methanol and 300 °C for DME), the experimental practice demonstrates that the measured composition well fits with the equilibrium composition calculated for $T_{EQ} = T_{RF}$. For this reason, for T_{RF} higher than 250 °C for methanol and 300 °C for DME the equilibrium temperature T_{EQ} has been assumed equal to the reforming temperature. In contrast, at low reforming temperatures (below 250 °C for methanol and 300 °C for DME), the reforming reactions are kinetically limited, especially at high pressures and with low SCRs. In this case, the composition of the reformed gas can

be again evaluated under the hypothesis of chemical equilibrium, even though the equilibrium temperature is lower than the reforming temperature. For this reason, a suitable equilibrium approach temperature $\Delta T_{EQ} = (T_{RF} - T_{EQ})$ has been considered here. No detailed information about the equilibrium approach temperature for methanol and DME are available in the literature. According to the correlation available for methane [27], the following correlations have been here considered for methanol and DME, respectively:

$$\Delta T_{EQ} = T_{RF} - T_{EQ} = \frac{250 - T_{RF}}{2.5} \qquad (11)$$

$$\Delta T_{EQ} = T_{RF} - T_{EQ} = \frac{300 - T_{RF}}{2.5} \qquad (12)$$

For both internally and externally reformed systems, the reformed fuel gas (composed of H_2, CO, CO_2, H_2O and unconverted fuel) flows through the fuel cell anode (An), where the hydrogen reacts with the oxygen ions coming from the cathode (Ct). The only electrochemical reaction considered here is H_2 oxidation owing to its higher reaction rate with respect to CO oxidation. However, even the CO participates indirectly in the anode reactions, as it is converted to H_2 by means of the CO shift reaction [2]. Therefore, the overall anode reactions are:

$$H_2 + \frac{1}{2}O_2 \rightarrow H_2O \qquad (13)$$

$$CO + H_2O \rightarrow CO_2 + H_2 \qquad (14)$$

Anode and cathode exhaust gases are mixed together and fed to the post-combustor (PC) to complete fuel conversion.

The current produced by the fuel cell is closely related to the hydrogen molar flow consumed by the anode reactions. Since hydrogen from the fuel and oxygen from the air are not completely consumed by the electrochemical reactions, the fuel mass flow at the anode inlet and air mass flow at the cathode inlet can be determined from the fuel utilization factor U_F (commonly in the range 60–90%) and the air utilization factor U_A (usually between 15 and 30%):

$$U_F = \frac{(m_{H_2})_{Consumed}}{(m_{H_2} + m_{CO})_{Anode\ inlet}} \qquad (15)$$

$$U_A = \frac{(m_{O_2})_{Consumed}}{(m_{O_2})_{Cathode\ inlet}} \qquad (16)$$

Power output and stack efficiency strongly depend on cell voltage V and current density J (their product gives the power density). Common voltage values for tubular SOFC stacks range from 0.5 to 0.8 V, whereas current density range usually falls between 100 and 500 mA cm^{-2} [1–2]. However, actual voltage V depends on fuel cell type and on the stack operating parameters (current density, operating

pressure and temperature, oxidant and fuel composition, air and fuel utilization factors, etc.). The stack model considered here requires as input data the cell voltage produced at reference operating conditions. Starting from this reference voltage, the model uses correlations from published data [1–2] to evaluate the voltage variation produced by a change of the operating parameters.

The energy balance of the stack requires that a suitable portion of the heat produced by the anode and post-combustor reactions is directly recovered by the air pre-heater (AH) and, if present, the internal fuel reformer. Stack heat losses (Q_{LOSS}) are evaluated as a given fraction of fuel cell energy input. For given values of the stack operating parameters, composition and mass flows of the different streams are then calculated by means of the previous equations (13–16) and of the overall energy balance.

The energy balance of the external reforming section allows to calculate the mass flow and the exit temperature of the flue gas under the constraint of the minimum temperature difference ΔT_{RF}. Similarly, the energy balance of the recuperator gives both the air and the flue gas exit temperature, under the constraint of its minimum temperature difference ΔT_{RC}. Finally, the energy balances of the fuel vaporizer and the fuel heat exchanger allow to calculate the flue gas exit temperature, even according to their minimum temperature difference.

The analysis of hybrid SOFC-MGT power plants has been carried out with reference of a 100–150 kW-class methane fuelled MGT unit, with an air mass flow rate of 1 kg s^{-1} at the compressor inlet. Table 1 shows the main assumptions for the performance assessment of the externally and internally reformed SOFC-MGT hybrid power plants examined here.

3 Results and discussion

As already pointed out by the authors in other studies, methanol, ethanol and DME can be very interesting fuels for SOFC-MGT power plants [15–17]. In particular, Figs. 4 and 5 show efficiency and power output versus fuel utilization factor U_F for the internally reformed hybrid SOFC-MGT power plant fuelled by different fuels. For a given U_F, Figs. 4 and 5 demonstrate that simply replacing methane with methanol, ethanol or DME in SOFC-MGT systems slightly reduces efficiency (by about 3.0 points for DME, 1.0 point for ethanol and 0.5–2.0 points for methanol, depending on U_F) and power output (by about 16–18% for DME, 14–15% for ethanol and 12–22% for methanol, depending on U_F). For all fuels, increasing U_F always improves the performance because of higher SOFC fuel conversion efficiency. Moreover, ethanol and DME behave

**Table 1** Main operating parameters of the SOFC-MGT hybrid power plant

SOFC		MGT	
Methane LHV	50 MJ kg^{-1}	Air mass flow	1 kg s^{-1}
Methanol LHV	19.9 MJ kg^{-1}	Compressor inlet pressure drop	1.0%
Ethanol LHV	26.8 MJ kg^{-1}	Pressure ratio	4
DME LHV	28.4 MJ kg^{-1}	Turbine inlet temperature	950 °C
Stack current density	300 mA cm^{-2}	Compressor polytropic efficiency	82.0%
Stack operating temperature	900 °C	Turbine polytropic efficiency	82.0%
Methane steam-to-carbon ratio	2.0	Minimum RC and FH temp. differ.	20 °C
Methanol steam-to-carbon ratio	1.0	Minimum FV exit temperature	120 °C
Ethanol steam-to-carbon ratio	1.5	RC pressure drop (air side)	3.0%
DME steam-to-carbon ratio	1.5	FH fuel exit temperature	300 °C
Pressure drop (% of inlet pressure)	4.0%	FH and RC (gas side) pressure drop	5.0%
Heat losses (% of fuel energy input)	3.0%	FV pressure drop (gas side)	1.5%
DC/AC conversion efficiency	95.0%	Generator effic. (incl. aux. Cons.)	89.0%

in a very similar manner to methane, unlike methanol which behaves quite differently.

For a given U_F, the inferior performance of the alternative fuels can be attributed mainly to the different composition of the reformed fuel gas and to the lower heat required by the internal fuel reforming process. As a matter of fact, cell voltage (and stack efficiency) mainly depends on the average partial pressure of hydrogen, water and oxygen inside the anode and the cathode. Substituting methanol for methane does not produce a significant difference in cell voltage, whereas ethanol and DME reduce cell voltage (and stack efficiency). Obviously, the use of DME and ethanol also reduces stack power density, current density being constant. Moreover, for a given thermal input, methane reforming is more endothermic than ethanol, DME and methanol reforming. In particular, according to Eqs. 1–10 and to the different LHVs, for the stoichiometric SCRs considered here reforming heat is about

20.6% of the LHV for methane, about 14% for ethanol, 9.3% for DME and only 7.7% for methanol. For a given stack energy input, more heat needs to be removed by the air to keep the stack and turbine inlet temperatures constant, thus increasing air mass flow. Obviously, the increase of the air mass flow increases the fraction of the primary fuel energy converted only by the bottoming MGT plant instead of the combined SOFC-MGT system.

Methanol reforming is almost complete (that is fuel conversion is higher than about 99%) above 200–250 °C and DME reforming above 250–300 °C, whereas the complete reforming of methane and ethanol requires higher temperatures (700–900 °C) [19–26]. As the MGT exhaust gas is available at about 250–300 °C, the lower reforming temperature of methanol and DME allows siting at the reformer outside the stack, thus improving low temperature exhaust heat recovery. In contrast, methane and ethanol are unsuitable for externally reformed SOFC-MGT systems.
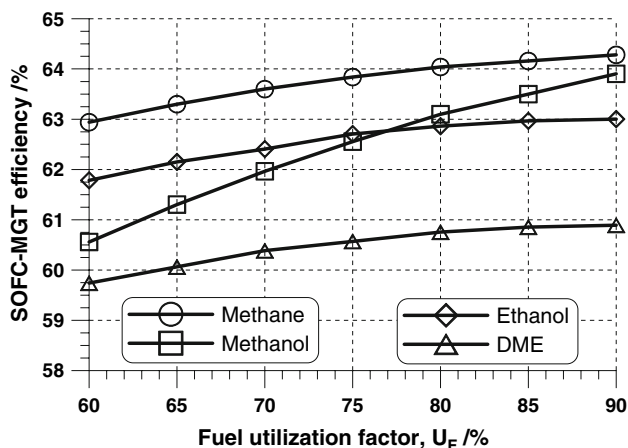
Fig. 4 Efficiency of internally reformed SOFC-MGT power plants fuelled by methane, methanol, ethanol and DME versus U_F
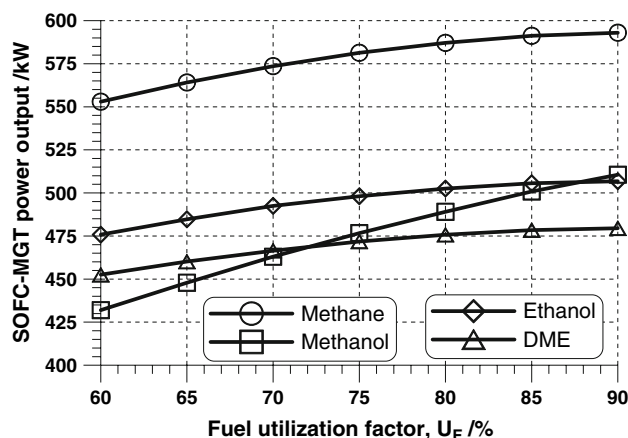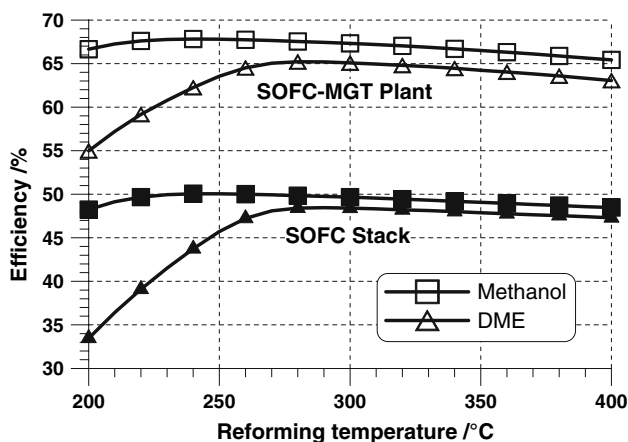

Fig. 5 Power output of internally reformed SOFC-MGT power plants fuelled by methane, methanol, ethanol and DME versus U_F

One of the most important operating parameters of externally reformed SOFC-MGT hybrid plants is the reforming temperature T_{RF}. Increase in the reforming temperature favours fuel conversion and the exhaust heat recovery. However, once fuel conversion reaches completion a further increase in T_{RF} requires higher temperatures of the reformer inlet gas and thus impairs the recuperator exhaust heat recovery.

Figures 6 and 7 show efficiency and power output versus reforming temperature of the SOFC stack and hybrid SOFC-MGT power plant fuelled by methanol and DME, for $U_F = 0,85$ and for the stoichiometric SCR values (1 for methanol and 1.5 for DME). Under the assumptions adopted here, Fig. 6 shows that methanol performs better than DME (the maximum hybrid plant efficiency is about 67% for methanol and 65% for DME) and that the best efficiency is achieved at reforming temperatures around 240–250 °C for methanol and 290–300 °C for DME. As shown by Fig. 7, stack and hybrid plant power output increase with T_{RF}. Moreover, DME gives higher power outputs, but only at reforming temperatures above 270–280 °C. As can be seen from Figs. 6 and 7, the performance of both stack and hybrid plant show very similar trends, as the performance of the MGT are practically independent of both primary fuel composition and reforming temperature.

As mentioned, the performance of the SOFC-MGT hybrid plants also depends on the SCR value. In fact, increasing SCR improves fuel conversion, but too high SCRs impair stack performance due to the lower hydrogen concentration produced by water dilution. Overall, the best hybrid plant efficiencies could be attained with SCRs slightly lower than the stoichiometric values. However, it must be observed that too small SCRs can lead to carbon formation [17].
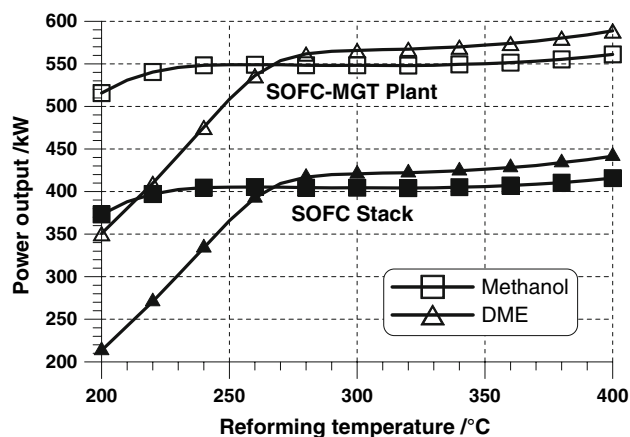
Fig. 7 Stack and plant power output of externally reformed SOFC-MGT power plants fuelled by methanol and DME versus T_{RF}

Figures 8 and 9 compare efficiency and power output of the internally reformed SOFC-MGT plant fuelled by methane and of the externally reformed SOFC-MGT power plant fuelled by methanol and DME. In particular, according to the optimum values of T_{RF} of Fig. 6, for methanol and DME two different reforming temperatures have been considered (250 and 300 °C).

As shown by Fig. 8, the efficiency of externally reformed SOFC-MGT power plants is higher that that of the internally reformed methane-fuelled plant only for U_F higher than 67–69% for methanol and 79–80% for DME. For given values of U_F and T_{RF}, methanol performs better than DME. The bottom part of Fig. 8 shows that for the most common values of U_F (below 85%), the stack efficiency of the externally reformed configurations is lower than that of the methane fuelled hybrid plant. Comparison of stack and hybrid plant efficiencies demonstrates that external reforming of methanol and DME improves hybrid plant efficiency owing to the increase in MGT exhaust heat recovery. As Fig. 9 shows, substituting external for internal reforming reduces stack and SOFC-MGT power output. Overall, the use of DME with $T_{RF} = 300$ °C yields higher power output, whereas the power output of the methanol fuelled SOFC-MGT power plant does not practically depend on T_{RF}.

Figure 10 compares anode H_2-to-H_2O partial pressure ratio (average value between anode inlet and anode outlet) for internal reforming of methane and external reforming of methanol and DME. For methanol and DME, the partial pressure ratio is very similar owing to the very similar reformed gas composition. Moreover, for both fuels external reforming significantly increases the hydrogen concentration and then partial pressure ratio. In fact, with external reforming anode gas recirculation is not required and the hydrogen is not diluted with the anode exhaust gas.
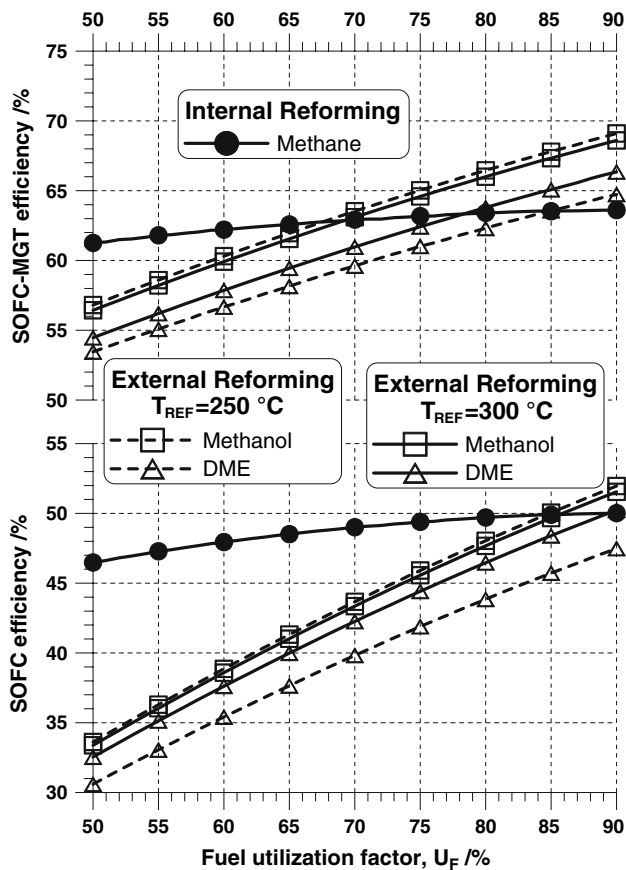

Fig. 6 Stack and plant efficiency of externally reformed SOFC-MGT power plants fuelled by methanol and DME versus T_{RF}

Fig. 8 Stack and plant efficiency of internally and externally reformed SOFC-MGT power plants versus U_F

Fig. 9 Stack and plant power output of internally and externally reformed SOFC-MGT power plants versus U_F

Fig. 10 Average partial pressure ratio H_2/H_2O of internally and externally reformed SOFC stacks versus U_F

What is more, during the external reforming process the CO shift reaction reaches equilibrium at a lower temperature (250–300 °C instead of 900 °C). Since this reaction is favoured by low temperatures, the hydrogen mole fraction of the reformed fuel gas produced by external reforming is very high (about 65–67%). On the other hand, the absence of a suitable catalyst inhibits the reverse CO shift reaction when the temperature increases from the stack inlet to the anode inlet [28].

As already mentioned, with external reforming the SOFC stack is fed directly by the reformed fuel gas, so the heat produced by the electrochemical and combustion reactions must be completely removed by the air. However, external reforming also requires a greater portion of the flue gas exiting the turbine to be sent to the reforming section, with a corresponding lower mass flow available for the recuperator and hence a lower air temperature at the stack inlet. Overall, external reforming of methanol and DME significantly increases the air cooling requirement of the SOFC stack and, as shown by Fig. 11, reduces the air utilization factor with respect to internal reforming of methane by about 5–10 points.
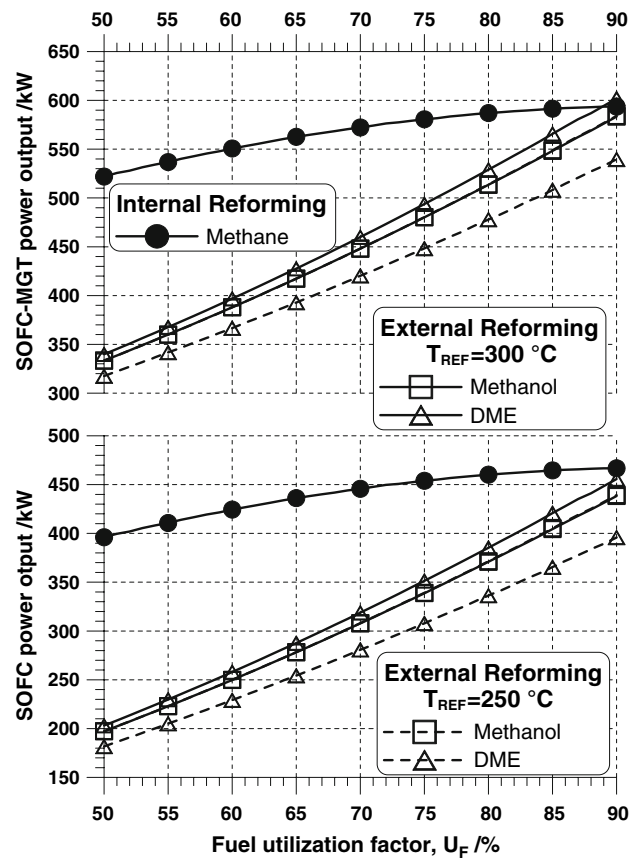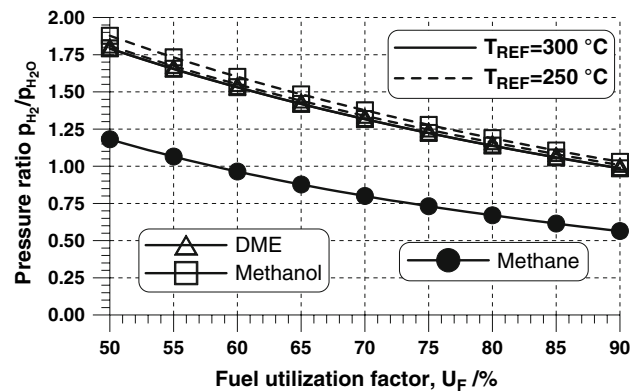
Figure 12 shows the substantial increase in cell voltage produced by external reforming of methanol and DME, due to the higher hydrogen partial pressure. Depending on U_F, the increase in cell voltage ranges from 75 to 100 mV, the highest values for methanol with T_{RF} = 250 °C. Obviously, external reforming also enhances stack power density, current density being constant.
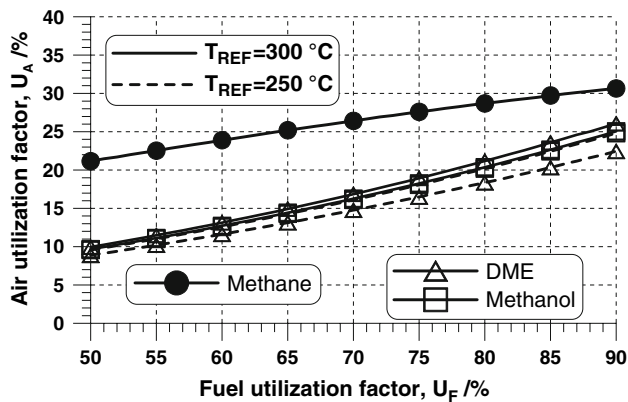
Fig. 11 Air utilization factor of internally and externally reformed SOFC stacks versus U_F
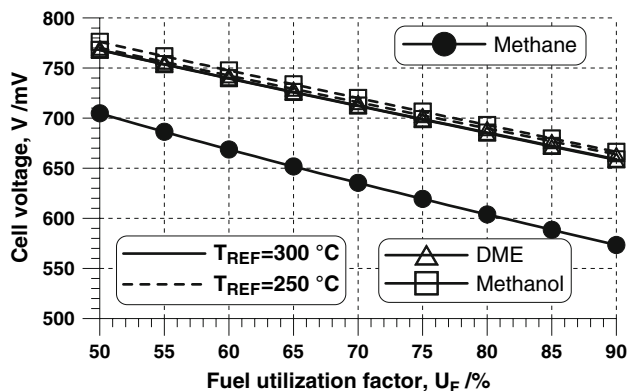
Fig. 12 Cell voltage of internally and externally reformed SOFC stacks versus U_F

4 Conclusions

Methanol, ethanol and DME are very attractive fuels for SOFC-MGT hybrid power plants. However, simply replacing methane with methanol, ethanol or DME in internally reformed SOFC-MGT systems slightly reduces efficiency and power output. On the other hand, the lower reforming temperature of methanol and DME (250–300 °C) allows siting at the reformer outside the stack, thus improving low temperature exhaust heat recovery. In contrast, methane and ethanol are unsuitable for externally reformed SOFC-MGT systems.

The performance assessment carried out in this paper has demonstrated that the operating parameters of the fuel reforming section (temperature and SCR) must be carefully chosen in order to optimise the performance of externally reformed SOFC-MGT hybrid plants. In particular, the optimum reforming temperature is around 240 °C for methanol (with hybrid plant efficiencies of about 67%) and

around 290 °C for DME (with hybrid plant efficiencies of about 65%).

Using methanol and DME in externally reformed SOFC-MGT hybrid plants can lead to efficiency improvement with respect to methane fuelled hybrid plants, especially for higher values of the fuel utilization factor. In particular, under the assumptions adopted here, the efficiency of externally reformed hybrid plants is higher that that of the internally reformed methane-fuelled plant for U_F higher than 67–69% for methanol and 79–80% for DME. External reforming also enhances efficiency on account of improved exhaust waste heat recovery and of the higher cell voltage produced by the greater hydrogen partial pressure at the anode inlet.

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