Short communication

Experimental results with a natural gas cogeneration system using a polymer exchange membrane fuel cell

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Abstract

This paper reports experimental results of an investigation of five identical CHP (combined heat and power) units using PEMFC (proton exchange membrane fuel cell) and running on natural gas. The natural gas is reformed locally to produce hydrogen. The net electric power is 4.5 kWe and the installations are designed for low temperature heat recovery (6 kW at 60 °C). The performances of the CHP units are analyzed in terms of electrical, thermal and total efficiencies. The electrical efficiency is low and it is shown that this is due mostly to the reforming process and to electric losses, while fuel cell performances are fully satisfying.

1. Introduction

The results we present refer to an electrical power system of 4.5 kWe, using PEMFC (proton exchange membrane fuel cell), running on natural gas and adapted to low temperature heat recovery (6 kW at 60 °C). It is designed and built by H-Power (RCU 4500 V2). Five identical units were put in operation in France between November 2002 and June 2003. The cities participating to these experiments in real operating conditions are Dunkerque (2 units), Nancy, Limoges and Sophia-Antipolis. This work was carried out within the framework of EPACOP project, led by *Gaz de France* and co-funded by the French agency for energy and environment (ADEME).

One of the most important characteristics of PEM fuel cells is their low operating temperature (50–80 °C), which is a drawback for efficient heat recovery [1].

2. System description and operation

Lombard et al. [2] published a detailed experimental analysis of the steam reforming unit (Fig. 1). The steam reforming and water–gas shift reactions occur at 650 °C in the reformer. Steam is fed in excess in order to inhibit amorphous coke formation [3]. The steam to carbon ratio S/C is between 6.5 and 8.1 [2], which is higher than the usual S/C ratio of natural gas conversion units for fuel cell applications [4]. Consequently, the water concentration of the outlet gas is high: between 0.35 and 0.55 mol_{H_2O}/mol_{gas} [2,5].

The cooler-shift eliminates by oxidation most of the carbon monoxide remaining in the reformer outlet gas. Although depending on the gas mixture flow rate, the conversion of carbon monoxide is good: 99.4% at $I = 40$ A and 98.6% at $I = 80$ A [2]. The gas is also cooled in two stages: first, high temperature heat (190–220 °C) is recovered by the steam; second, the cogeneration water further cools the gas through a heat exchanger located in the bottom part of the cooler-shift.

The prox is a catalytic reactor that eliminates the remaining carbon monoxide by preferential oxidation in the presence of a

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Nomenclature

E	fuel cell potential (V)
F	Faraday constant $F = 96485\,C\,mol^{-1}$
$\bar{g}$	Gibbs free energy ($J\,mol^{-1}$)
$\bar{h}$	enthalpy ($J\,mol^{-1}$)
I	current intensity (A)
N	number of cells in the fuel cell stack (–)
$\dot{n}$	molar flow rate ($mol\,s^{-1}$)
$\dot{Q}$	thermal power of the system (kW)
T_{FC}	fuel cell temperature (°C)
$\dot{W}_e^{gross/net}$	gross/net electric power of the system (kW)
x	maximum number of hydrogen moles produced by mole of natural gas
η	efficiency (–)
λ_{NG}	coefficient of excess of reformed natural gas: $\lambda_{NG} = \dfrac{\dot{n}_{NG}^{ref}}{\dot{n}_{NG}^{min}}$ (–)

Subscripts and superscripts

br	burned
comb	combustion
e	electrical
FC	fuel cell
NG	natural gas
ref	reformed
th	thermal

The fuel cell stack is made of $N = 120$ cells. Anode off-gas is injected into the reformer burner where excess hydrogen and the small amount of remaining methane are burned. An enthalpy wheel is used to recover water and heat from cathode outlet airflow and to transfer them to the cold and dry inlet airflow.

The primary water-cooling circuit goes successively through the shift, prox, and fuel cell. Then, heat is transferred to the user's circuit through another heat exchanger (not represented in Fig. 1). The maximum water temperature in the user's circuit is between 57 and 59 °C. If the demand is insufficient, heat is evacuated to the outside.

The units have an hybrid architecture: both fuel cell and batteries provide electricity. They integrate three electric converters: a DC/DC converter for raising the stack potential and making possible batteries charging, a DC/AC converter (60 Hz) for supplying the auxiliaries, and another DC/AC converter (50 Hz) for the main supply.

3. System efficiencies

3.1. Theoretical maximum electrical efficiencies

The maximum electrical efficiency of a fuel cell consuming hydrogen is given by (1)

$$\eta_{FC,H_2}^{max} = \frac{\Delta\bar{g}_{H_2,comb}}{\Delta\bar{h}_{H_2,comb}} \tag{1}$$

where $\Delta\bar{h}$ and $\Delta\bar{g}$ are the enthalpy and the Gibbs free energy of the overall reaction. The value of this ratio depends on temperature and activity of the reactants (H_2 and O_2) and of the product (H_2O): in the particular case of a PEMFC fed by hydrogen and air and operating at 60 °C, the theoretical maximum electrical efficiency equals 79% (since the cogeneration system recovers part of the latent heat of condensation of water, the HHV is used

small amount of air. The prox intake airflow is constant (470 *l/h*). Part of the oxygen reacts with carbon monoxide and the rest reacts with hydrogen. The amount of hydrogen consumed in the prox depends on the reformate flow rate (4.8% at $I = 40$ A and 3.1% at $I = 80$ A, [2]).
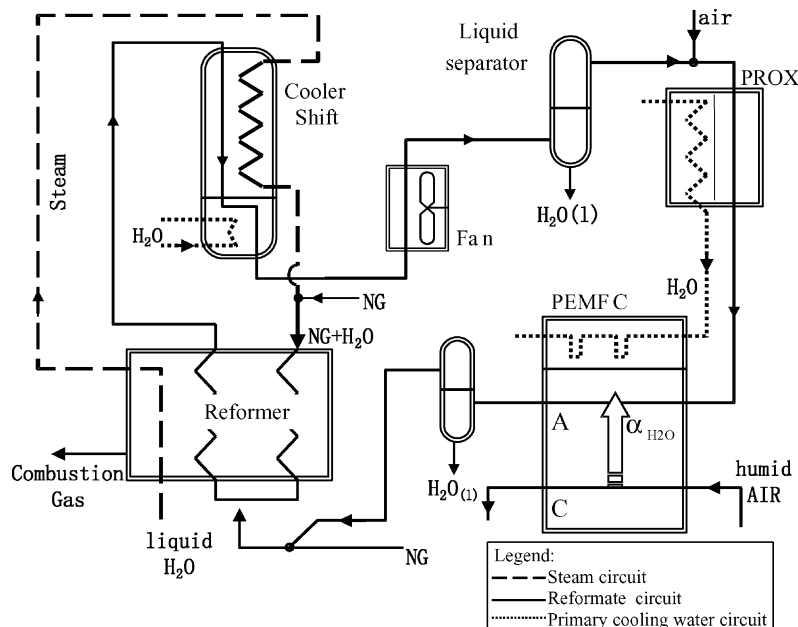

Fig. 1. System main components.

as reference for the determination of the efficiencies). However, in the units under consideration, heat must be supplied for the reforming of natural gas; assuming the system isothermal and isobar, the theoretical maximum electrical efficiency is given by (2)

$$\eta_{NG \to H_2 \to \bar{e}}^{max} = \frac{x \cdot \Delta \bar{g}_{H_2,comb}}{\Delta \bar{h}_{NG,comb} - \Delta \bar{h}_{NG,ref}} \qquad (2)$$

The molar flow rate of *reformed* natural gas is used as the reference for establishing Eq. (2). The numerator represents the electrical energy produced per mole of *reformed* natural gas; x is the number of moles of hydrogen produced per mole of *reformed* natural gas. Assuming complete steam reforming and water–gas shift reactions, x is given by (3)

$$x = \frac{\dot{n}_{H_2}}{\dot{n}_{NG}^{ref}} = 4 \cdot c_{CH_4} + 7 \cdot c_{C_2H_6} + 10 \cdot c_{C_3H_8} \qquad (3)$$

where c_{CH_4}, $c_{C_2H_6}$ and $c_{C_3H_8}$ are the natural gas mole fraction of methane, ethane and propane, respectively. In the denominator of Eq. (2), $\Delta \bar{h}_{NG,ref}$ stands for the enthalpy of reforming reactions and $\Delta \bar{h}_{NG,comb}$ is the enthalpy of the reaction of combustion of natural gas. $\Delta \bar{h}_{NG,ref}$ is provided by the combustion of natural gas and anode off-gas. One can check easily that $\Delta \bar{h}_{NG,comb} - \Delta \bar{h}_{NG,ref} = x \cdot \Delta \bar{h}_{H_2,comb}$, which finally yields (4):

$$\eta_{NG \to H_2 \to \bar{e}}^{max} = \eta_{FC,H_2}^{max} \qquad (4)$$

Therefore, from a thermodynamic point of view, the electrical efficiency of an electro-chemical system using PEMFC is the same whether hydrogen is supplied directly or produced locally by reforming of natural gas.

3.2. Actual efficiencies

The fuel cell effective electrical efficiency is the ratio between the stack electric power and the enthalpy flow rate associated with the reaction of hydrogen combustion, which simplifies into (5):

$$\eta_{FC} = \frac{E}{HHV_{H_2} \cdot N/2F} \qquad (5)$$

The effective electrical efficiency of the cogeneration system (6) is the ratio between the electric power and the enthalpy flow rate associated with the reaction of combustion of natural gas:

$$\eta_{system}^{e,gross} = \frac{\dot{W}_e^{gross}}{\dot{n}_{NG} \cdot HHV_{NG}}, \qquad \eta_{system}^{e,net} = \frac{\dot{W}_e^{net}}{\dot{n}_{NG} \cdot HHV_{NG}} \qquad (6)$$

where $\dot{n}_{NG}$ is the total molar flow rate of natural gas fed to the system. $\dot{n}_{NG}$ is the sum of the amounts of natural gas that are reformed $\dot{n}_{NG}^{ref}$ and burned $\dot{n}_{NG}^{br}$. The net electric power available to the user is deduced from the stack electric power $EI = \dot{W}_e^{gross}$ by subtracting the electric consumption of the auxiliaries and the losses in the electric converters (which were experimentally determined).

The gross electrical efficiency can also be written as the product of fuel cell and reforming effective efficiencies $\eta_{system}^{e,gross} =$

$\eta_{FC} \cdot \eta_{ref}$. Following Lutz et al. [6] and Mathiak et al. [7], the reforming efficiency η_{ref} is defined as the ratio between the enthalpy flow rates of hydrogen and of natural gas (7):

$$\eta_{ref} = \frac{I \cdot N/2F \cdot HHV_{H_2}}{\dot{n}_{NG} \cdot HHV_{NG}} \qquad (7)$$

Note that this definition (7) is based on the hydrogen and natural gas flow rate *consumed* by the system or by the fuel cell. One should consider Eq. (7) as the efficiency of the reforming process rather than the efficiency of the reforming unit (in that case, the hydrogen flow rate *supplied* to the fuel cell should appear in the numerator).

The system thermal efficiency is the ratio between the useful thermal power and the enthalpy flow rate associated with the reaction of combustion of natural gas (8):

$$\eta_{system}^{th} = \frac{\dot{Q}}{\dot{n}_{NG} \cdot HHV_{NG}} \qquad (8)$$

The system thermal power $\dot{Q}$ is the sum of the heat fluxes recovered at the cooler-shift, at the prox and at the fuel cell. The net total efficiency is the sum of the net electrical efficiency and of the thermal efficiency.

4. Experimental results

Data collected from the 5 units were used to determine their actual electric, thermal and total efficiencies. The results refer either to instantaneous values recorded each 5 min, to stable operating points, or to values averaged over continuous periods of operation ranging from 2 days to one month. An operating point is considered stable when the current intensity remains constant during at least 30 min.

4.1. Fuel cell electrical efficiencies

Starting from stable operating points, a regression of fuel cell polarization curve shows that it is well approximated by a linear Eq. (9):

$$E_{FC} = a - b \cdot I \qquad (9)$$

with $a = 96.7$ V and $b = 0.27 \, \Omega$. The maximum absolute deviation between the points and the line is 4.5 V while the mean absolute deviation is 0.74 V. The active area of a single cell is 217 cm^2 and the maximum current density is 4610 A/m^2 at $I = 100$ A. The fuel cell electrical efficiency varies between 47%$_{HHV}$ at $I = 100$ A (58% of the thermodynamic efficiency) and 63%$_{HHV}$ at $I = 15$ A (79% of the thermodynamic efficiency). A statistical analysis of instantaneous operating points recorded on Limoges' unit between November 20th and December 4th, 2003, indicates that 56.2% of them are above the regression line corresponding to steady state operation (Fig. 2). Furthermore, the relative deviation from this line is less than 2% for 93.1% of them: one can conclude that (in this system) the electrical efficiency of the fuel cell is not sensitive to transient operation.
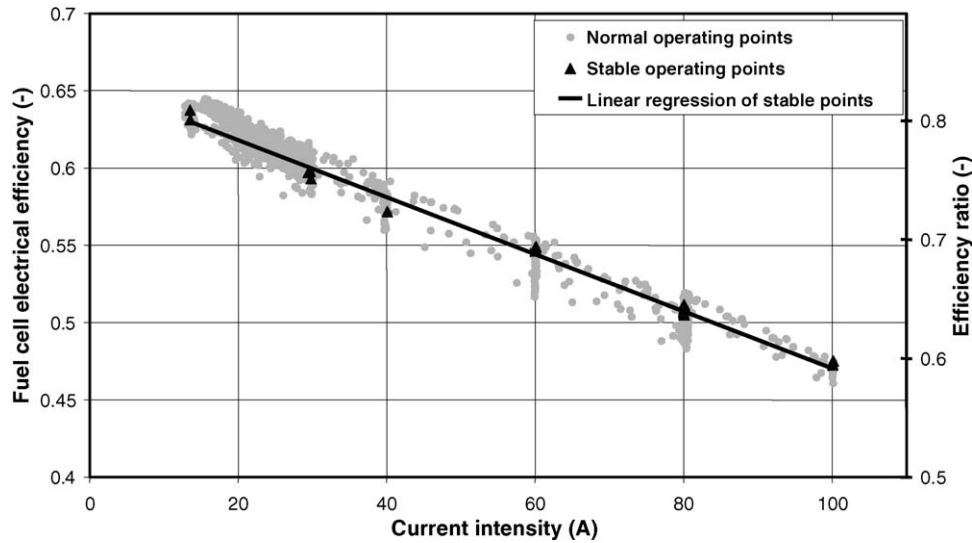
Fig. 2. Fuel cell electrical efficiency η_{FC} (Eq. (5), left vertical axis) and ratio to maximum efficiency ($\eta_{FC}/\eta_{FC}^{max}$, right vertical axis) measured on Limoges' unit between November 20th and December 4th, 2003.
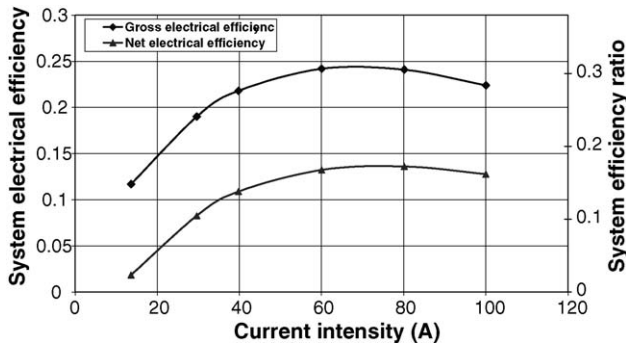

Fig. 3. System gross and net electrical efficiencies $\eta_{system}^{e,gross}$ and $\eta_{system}^{e,net}$ (Eq. (6) and left vertical axis) and ratio to maximum efficiency ($\eta_{system}^{e,gross/net}/\eta_{FC,H_2}^{max}$, right vertical axis) in stable operation.

Fig. 4. Measured reformed natural gas excess λ_{NG} and reforming efficiency η_{ref} as functions of current intensity.

4.2. System electrical efficiency

Fig. 3 shows the evolutions of the system gross and net electrical efficiencies $\eta_{system}^{gross/net}$ (6) as functions of current intensity. The values are low compared to the fuel cell stack electrical efficiency, all the more so considering that the theoretical maximum values are the same in both cases (4): according to Fig. 3, the actual values of the system gross electrical efficiency (6) are between 12 and 30% of the maximum thermodynamic efficiency[1] (4). It must also be noted that the behavior of the curves are different: the stack electrical efficiency is a decreasing function of the current intensity while the gross and net electrical efficiencies reach an optimum between $I = 60$ and 80 A. Fig. 3 also shows that the difference between gross and net electric powers is high and results in a quite important degradation of the system net efficiency, which becomes very low or even nil at
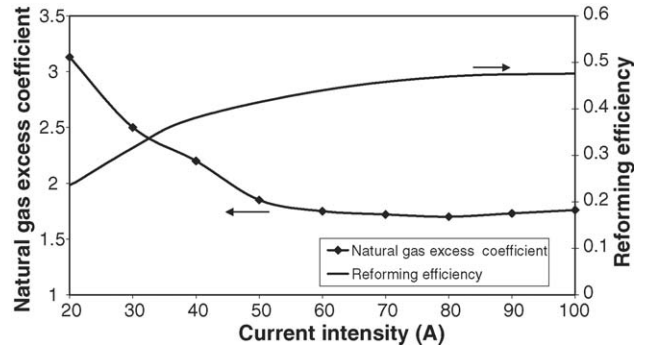
the lowest intensities. The complexity of the electric architecture of the system, which integrates 3 electric converters (Section 2), is at the origin of these high electric losses[2]. Another statistical analysis of instantaneous operating points recorded on Limoges' unit between November 20th and December 4th, 2003, indicates that for most of them (84.5%) the system gross electrical efficiency is below the steady state curve. The supplementary loss of electrical efficiency due to transient operation is estimated at about 1.5 percentage points. This corresponds to a relative loss of electric power of about 6%.

Fig. 4 shows the evolution of the coefficient of excess of reformed natural gas λ_{NG} as a function of the current intensity; λ_{NG} stands for the ratio between the amount of natural gas supplied to the reformer $\dot{n}_{NG}^{ref}$ and the minimal quantity theoretically required $\dot{n}_{NG}^{min} = x \cdot I \cdot N/2F$, where x is given by Eq. (3). λ_{NG} decreases with the current intensity. It is never lower than 1.7 and can be above 3 at $I = 20$ A. One can also observe in Fig. 4 (on right axis) that the reforming efficiency η_{ref} (7) increases with intensity, following an opposite trend to that of λ_{NG}: high

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[1] These results confirm those of Gigliucci et al. [5]. However, they refer only to the H-Power RCU 4500 V2 units and should not be considered as representative of the current Plug Power products, such as GenSys™, which are more advanced and which operate in different conditions [8].

[2] Electric losses (including inverters and auxiliaries consumption) can by evaluated by: $0.7\,kW + 0.3\dot{W}_e^{gross}$.

values of λ_{NG} increase the reformer vapor consumption, which is detrimental to the reforming efficiency. Lombard et al. [2] showed that the reformate composition can be fairly approximated assuming thermodynamic equilibrium at the reformer outlet. This makes it possible to evaluate accurately the reforming efficiency as a function of *S/C* ratio, λ_{NG}, and burner exhaust gas temperature T_{ex}. With *S/C* = 6, λ_{NG} = 1.2 and T_{ex} = 150 °C, the result is η_{ref} = 0.55. This is probably the best value achievable with this reforming unit: it would lead to a system gross electric efficiency ranging between 26%$_{HHV}$ and 35%$_{HHV}$. By comparison, in the 10–30 A range *S/C* is above 8, λ_{NG} = 2.5–3.1, $T_{ex} \cong 200$ °C and as a consequence, η_{ref} = 0.2–0.3.

4.3. System thermal and total efficiencies

Electric and energy meters allow evaluating mean values of the net electrical, thermal and total efficiencies over periods of operation ranging from a few days to a full month. These values take account of all the thermal and electric losses, including start up time (90 min) during which the units are supplied with natural gas without producing electricity. An analysis of all data recorded during more than one year shows that the net electrical, thermal, and total efficiencies are very low. The global mean value of current intensity (averaged for all units and weighted by the length of the operation periods) is 36 A, which is quite far from the optimum range (60–80 A, Fig. 2). As a consequence, the global mean electrical efficiency is only 9.2%. The global mean value of thermal efficiency is 29%, meaning that heat recovery must be improved drastically.

5. Conclusions and perspectives

Although the operation of the fuel cell itself is fully satisfactory, the electrical and thermal performances of the tested cogeneration units are disappointing. The experimental data show that the poor electrical efficiency is due mostly to high electric losses and to the need of vaporizing excess water for the fuel-reforming process (worsened by the reforming of a large excess of natural gas). It is shown that in term of reforming efficiency, significant improvements are achievable.

It also appears that whatever the installation, the electricity demand is not adapted to the systems. All of them are used essentially at part load, far from their optimum. The poor thermal efficiency is due to the low temperature of the fuel cell and to the presence of a primary cooling circuit (the user's circuit should be used directly as a cooling circuit).

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