

Short communication

Shortcut model for water-balanced operation in fuel processor fuel cell systems

P. Maarten Biesheuvel*, Gert Jan Kramer

Shell Global Solutions International b.v., Badhuisweg 3, 1031 CM, Amsterdam, The Netherlands

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Abstract

In a fuel processor, a hydrocarbon or oxygenate fuel is catalytically converted into a mixture rich in hydrogen which can be fed to a fuel cell to generate electricity. In these fuel processor fuel cell systems (FPFCs), water is recovered from the exhaust gases and recycled back into the system. We present a simple mass balance analysis based on the assumption that the off-gas leaving the system is just saturated (perfect condenser). The model results in simple expressions for the net amount of water produced, and the critical condition for water-balanced operation in FPFCs. The analysis includes the composition of the hydrocarbon or oxygenate fuel, the air-to-fuel inlet ratio, humidity, ambient temperature and pressure, and the temperature and pressure in the condenser. The analysis can be used to quickly assess under what conditions operation is critical and additional measures or alternative water recovery technologies are required. The simple analysis is in agreement with the more extensive mass balance analysis by Ahmed et al. [Water balance in a polymer electrolyte fuel cell system, *J. Power Sources* 112 (2002) 519–530] and shows the same dependencies of the water balance on the H/C ratio, condenser pressure, ambient temperature, etc. The analysis shows that as long as these parameters remain invariant, the actual amount of water that is used in the system is of no importance from an overall water management perspective. For instance, high steam loads in the reformer, or in the fuel cell (e.g., in case of a polyelectrolyte membrane fuel cell) do not burden the overall water balance in the least.

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1. Introduction

In fuel processor fuel cell systems (FPFCs), a (hydrocarbon) fuel is catalytically converted with air to a mixture rich in hydrogen before being fed to a fuel cell to generate electricity [1–5]. FPFCs are envisioned as an important transition technology towards an established hydrogen economy in which hydrogen is generated from renewable sources, stored and transported, and used as fuel without requiring intermediate oxygenate or hydrocarbon fuels. FPFCs still use hydrocarbon or oxygenate fuels to generate hydrogen; subsequently, the

hydrogen can be fed to a fuel cell. Besides their relevance in facilitating the development of a hydrogen economy, FPFCs have significant potential in their own right, e.g., for small-scale electricity production from liquid or gaseous fuels at the scale of a single business, household or vehicle. Currently, FPFC technology is reaching beyond the pilot plant stage, but the high costs of catalysts and fuel cells are still a major limitation to a large breakthrough.

In this paper, we discuss one aspect of FPFC operation, namely related to the recovery of water from the off-gas and its subsequent re-use in the system to obtain water-balanced operation. This subject was recently discussed extensively and quite well by Ahmed et al. [1], to whom we will refer at several instances. Though several of our conclusions are quite similar to theirs, the objective of our paper is the presentation of a simple method to calculate the amount of excess

* Corresponding author. Present address: Laboratory of Physical Chemistry and Colloid Science, Wageningen University, Dreijenplein 6, 6703 HB Wageningen, The Netherlands. Tel.: +31 317 482 189; fax: +31 317 483 777.
E-mail address: maarten.biesheuvel@wur.nl (P.M. Biesheuvel).

Nomenclature

ELW	excess liquid water (mol/s)
ELW*	excess liquid water divided by C-atom flow ϕ_C (-)
RH	relative humidity (-)
P	pressure (bar)
p^*	reference pressure (bar)
p_i^{sat}	equilibrium vapour pressure at T_i (bar)
T	temperature (K)
T^*	reference temperature (K)
T^{CRIT}	maximum, critical, temperature in condenser (K)
y	fraction water in exhaust of condenser (-)
Y	hydrogen yield (-)
α_{TOT}	overall molecular O_2 flow, divided by atomic carbon flow, ϕ_C (-)
α_{REF}	molecular O_2 flow to reformer, divided by ϕ_C (-)
β	atomic H/C ratio of fuel (-)
γ	atomic O/C ratio of fuel (-)
ΔH_{vap}	enthalpy of vaporisation of water (J/mol)
ζ_1	fuel utilization in fuel cell (-)
ζ_2	oxygen utilization in fuel cell (-)
ϕ	molar flow (mol/s)
χ	fraction oxygen in air (=0.2095) (-)

Subscripts

A	air entering system
C	condenser (in case of P_C and T_C)
C	carbon (in case of ϕ_C)

water produced, and the critical condition for water-balanced operation. Such an analysis is useful because it clearly shows the key parameters in water management and can be used to quickly identify under which conditions additional measures are required for water-balanced operation (e.g., increase of condenser pressure, lowering of air intake) or whether alternative technologies (other than a condenser) need to be applied. In our analysis, we only incorporate the parameters that directly influence water management and neglect further details of the fuel reformer and fuel cell. For instance, we use the reformer air-to-fuel ratio α_{REF} as an input parameter and do not calculate what actually is an optimal value for α_{REF} or how the optimal value depends on the composition of the fuel. These issues require a full analysis of the entire FPFC system, which is beyond the objective of the shortcut model.

The off-gas from a FPFC is released to the atmosphere after a condensation step to recover (part of) the water, see Fig. 1. The recovered water is used for several purposes within the FPFC to make operation possible and improve system efficiency. In the lay-out of the technology we are most famil-

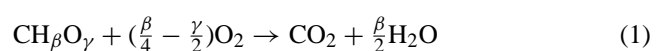
iar with [5,6], water is used besides air for the generation of synthesis gas (in so-called “autothermal reforming” or “catalytic partial oxidation”) to suppress high temperatures and increase efficiency, in the water–gas shift converter to obtain additional hydrogen, and finally to humidify the streams entering the polyelectrolyte membrane (PEM) fuel cell to keep the fuel cell fully humid.

For logistic reasons and due to the required purity, the water should preferentially be auto-generated by separation from the off-gas streams without requiring an external source of (de-ionized) water. The high required purity is related to the sensitivity of system components to contaminants and ions, as well as due to the inevitable accumulation of condensables in the system. Fresh, clean water can certainly be generated from available other water sources (e.g., tap water) but the (energy) costs of filtration and ion removal are very high. Such an additional technology would significantly reduce the overall efficiency of the FPFC (“parasitic loss”) because it is the electricity produced in the FPFC that must be used for the purification. It must also be noted that the water circulation rate can be very high and may be of the order of ~ 10 times the (liquid) fuel volume flow that enters the system. This again shows the relevance of water recovery.

To understand whether enough water is recovered from the exhaust we do not consider each unit within the FPFC separately, but set up a balance for the air and water flows across the entire FPFC and derive simple relations for the net amount of water produced (or consumed) as well as the critical condition for operation in a water-balanced mode. The relevant parameters in the model are the pressure and temperature in the condenser, the overall air-to-fuel ratio, the fuel hydrogen/carbon and oxygen/carbon ratio and ambient pressure, temperature and humidity. The only assumptions that are made in the model are (1) stationary operation, (2) off-gas leaving the condenser is (at the condenser temperature and pressure) just saturated and (3) full fuel and hydrogen combustion before the gas is fed to the condenser. It must be noted that given the above assumptions, the model is valid, irrespective of the details and type of fuel processor and fuel cell, as long as there is need for at least some recovery of water. Thus, as long as the relevant overall parameters of the system are invariant (fuel composition, air–fuel ratio, temperature and pressure of the air, and in the condenser), a system with high internal water flow rates (e.g., because the installed fuel cells require fully water-saturated gas-streams) is with respect to operation in a water-balanced mode completely equivalent to a technology that requires less water (e.g., based on solid-oxide fuel cells).

2. Theory

The overall reaction taking place in a FPFC is



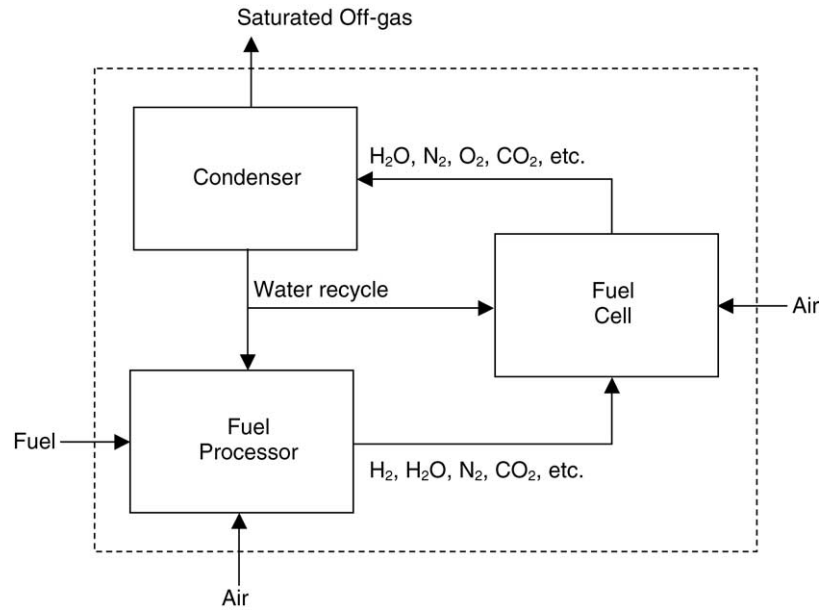


Fig. 1. Schematic overview of fuel processor-fuel cell system with condenser. The dashed line shows where we draw the boundary of the system.

with β the atomic H/C ratio of the fuel, and γ the atomic O/C ratio of the fuel (further on, α will be the inlet flow rate of O_2 molecules in the air divided by the inlet flow rate of fuel C-atoms). Eq. (1) can be used for all oxygenate and hydrocarbon ($\gamma = 0$) fuels as well as for pure hydrogen ($\gamma = 0$, $\beta \rightarrow \infty$). Eq. (1) is an overall balance and shows that fuels of high H/C ratio, β , produce more water while the amount of water produced per mole of carbon is independent of the oxygenation degree of the fuel, γ [1]. A higher γ can be advantageous for operation in a water-balanced mode due to the lower H_2 -yield Y in the reformer (see Eq. (8) further on), consequently a lowering of the cathode air flow to the fuel cell (at constant fuel and oxygen utilization), and therefore less water lost inadvertently with the condenser off-gas, as will be discussed in more detail in the next section.

The overall flow of oxygen, ϕ_{O_2} , into the system is $\alpha_{TOT} \cdot \phi_C$ with ϕ_C the inlet flow of C-atoms in the fuel and $\alpha_{TOT} > \beta/4 - \gamma/2$; the airflow ϕ_{air} is ϕ_{O_2}/χ with $\chi = 0.2095$. The dry exhaust gas flow (from the condenser) ϕ_{exh}^{dry} is given by

$$\phi_{exh}^{dry} = \phi_{air} + \phi_C \left(1 - \frac{\beta}{4} + \frac{\gamma}{2}\right) \quad (2)$$

The fraction of water in the exhaust of the condenser, y , is given by

$$y = \frac{p_C^{sat}}{P_C} = \frac{\phi_{exh}^{H_2O}}{\phi_{exh}^{H_2O} + \phi_{exh}^{dry}} \quad (3)$$

where p_i^{sat} is the equilibrium vapour pressure of water at T_i . P_C is the pressure in the condenser, and $\phi_{exh}^{H_2O}$ is the water flow in the exhaust gas leaving the system. In Eq. (3), we assume that the off-gas is just saturated. Because generally $\phi_{exh}^{H_2O} \ll \phi_{exh}^{dry}$, we will neglect $\phi_{exh}^{H_2O}$ in the denominator of

Eq. (3), which simplifies the further analysis significantly. Though the error made by making this assumption is small, an improved model can be constructed in a straightforward manner based on Eq. (3).

Assuming $\phi_{exh}^{H_2O} \ll \phi_{exh}^{dry}$, the water balance over the system is given by

$$\phi_{air} \cdot RH \cdot \frac{p_A^{sat}}{P_A} + \frac{\beta}{2} \phi_C = \phi_{exh}^{dry} \frac{p_C^{sat}}{P_C} + ELW \quad (4)$$

with the relative humidity, RH, used as a fraction (e.g., RH = 0.3 for 30% relative humidity), A referring to the air entering the system, and C referring to the condenser exhaust gas leaving the system. ELW is the excess liquid water produced (which can be temporarily stored or drained) and T_A , P_A , T_C and P_C are the temperature and pressure at ambient conditions and in the condenser, respectively. Combination of Eqs. (3) and (4) results after division by ϕ_C in the water balance over the entire FPFC system

$$\frac{\alpha_{TOT}}{\chi} RH \frac{p_A^{sat}}{P_A} + \frac{\beta}{2} = \left(\frac{\alpha_{TOT}}{\chi} + 1 - \frac{\beta}{4} + \frac{\gamma}{2} \right) \frac{p_C^{sat}}{P_C} + ELW^* \quad (5)$$

with $ELW^* = ELW/\phi_C$. Eq. (5) compares the intake of water (as humidity in the air) and the production of water from the fuel (first two terms) with the inadvertent loss of water in the condenser. The remainder is excess water produced in a useable, liquid, form (ELW^*), which can be stored, recycled or drained. With Eq. (5) we take a slightly different approach from Ahmed et al. [1] who (using the same system boundary) are more focused on analysing what is required for the separate elements in the system.

For p_i^{sat} we use ([7], p. 143)

$$p_i^{\text{sat}} = p^* \exp \left(-\frac{\Delta H_{\text{vap}}}{R} \left(\frac{1}{T_i} - \frac{1}{T^*} \right) \right) \quad (6)$$

with $T^* = 300$ K, $p^* = 35.4$ mbar (at T^*), $\Delta H_{\text{vap}} = 43.5$ kJ/mol (at T^*) and $R = 8.3144$ J/(mol K). An important assumption with respect to Eq. (6) is that thermodynamic equilibrium is reached in the condenser: condensation occurs sufficiently fast (no supersaturation) and the vapour droplets, which are formed are completely separated out from the gas-stream. In reality thermodynamic equilibrium will only be reached to a certain extent, thereby aggravating the water balance.

The overall air-to-fuel ratio, α_{TOT} , being the ratio of the total number of molecules of O_2 that enter the system in the air, over the number of C-atoms in the fuel, is a summation of the air intake in the different sections of the system, including the reformer, the fuel cell and a possible spent-gas burner installed prior to the condenser to remove traces of hydrocarbons and non-utilized hydrogen. Because practically all air is either added to the reformer or to the fuel cell (assuming that the spent-gas burner is fed with cathode air from the fuel cell) we obtain for α_{TOT}

$$\alpha_{\text{TOT}} \sim \alpha_{\text{REF}} + \frac{Y\zeta_1}{2\zeta_2} \quad (7)$$

with α_{REF} the (molecular) oxygen flow to the reformer (scaled to ϕ_C), with $\alpha_{\text{REF}} = 0$ for pure steam reforming. Y is the yield of hydrogen in the reformer, which is the number of moles of hydrogen produced in the reformer (that is, after the water-gas-shift reactor and a possible stage to remove remaining CO), per mole of C-atoms of the fuel [5]. In the fuel cell, ζ_1 is the fuel utilization and ζ_2 the oxygen utilization (one over the air stoichiometry). Ahmed et al. [1] use the following values for methane reforming: $\alpha_{\text{REF}} = 0.478$, $Y = 3.05$, $\zeta_1 = 0.8$ and $\zeta_2 = 0.4$ resulting in $\alpha_{\text{TOT}} = 3.53$. To obtain a relation between Y and α_{REF} , we use a similar approach as in Biesheuvel and Kramer [5]. For an oxygenate fuel of arbitrary composition the result for 100% fuel and 100% oxygen conversion is

$$Y = 2 + \frac{\beta}{2} - \gamma - 2\alpha_{\text{REF}} \quad (8)$$

which indeed gives $Y = 3.05$ for $\alpha_{\text{REF}} = 0.478$ (for methane, $\beta = 4$, $\gamma = 0$). Combining Eqs. (7) and (8) results in

$$\alpha_{\text{TOT}} \sim \alpha_{\text{REF}} \left(1 - \frac{\zeta_1}{\zeta_2} \right) + \left(1 + \frac{\beta}{4} - \frac{\gamma}{2} \right) \frac{\zeta_1}{\zeta_2} \quad (9)$$

Interestingly, because ζ_1/ζ_2 is generally >1 (e.g., $\zeta_1/\zeta_2 = 2$ for the data given above), Eq. (9) shows that a higher α_{REF} results in a lower α_{TOT} . This is due to the fact that a higher α_{REF} results in a lower yield of hydrogen and thus (for a given ζ_1 and ζ_2) a lower cathode air flow, which overrules the higher air flow to the reformer [1].

It must be noted that the above analysis (Eqs. (4), (5) and (7)) is based on a quite ideal line-up of the system, especially

with respect to using cathode air for the spent-gas burner and the location of the burner in front of the condenser. If it is placed after the condenser (attractive with respect to heat management and the reduced water load), significant amounts of water will travel through the condenser in the form of unconverted H_2 (and CH_4 remnants), will not condense out and thereby significantly deteriorate the water balance. However, when a second condenser is placed behind the burner, water recovery is again at a maximum (when cathode air is used as oxidant once again) and Eqs. (4), (5) and (7) can be directly applied.

Combination of Eqs. (5) and (6) gives the excess liquid water, ELW^* , as function of RH , T_A , P_A , P_C , T_C , β and γ . It is also possible to assume $\text{ELW}^* = 0$ and calculate the critical condition, e.g. the maximum, critical, temperature in the condenser, $T_C = T^{\text{CRIT}}$ as function of the other parameters. To make the dependence of T^{CRIT} on the other parameters explicit, we rewrite Eqs. (5) and (6) to obtain the critical condenser temperature (in K)

$$T^{\text{CRIT}} = \left\{ \frac{1}{T^*} - \frac{R}{\Delta H_{\text{vap}}} \ln \left(\frac{P_C (\alpha_{\text{TOT}}/\chi) \cdot \text{RH} \cdot (p_A^{\text{sat}}/P_A) + \beta/2}{p^* (1 + \alpha_{\text{TOT}}/\chi - \beta/4 + \gamma/2)} \right) \right\}^{-1} \quad (10)$$

with p_A^{sat} a direct function of T_A according to Eq. (6).

To simplify a bit, for a hydrocarbon fuel ($\gamma = 0$), $\text{RH} = 0$ and $\alpha_{\text{TOT}}/\chi \gg 1$ we obtain for the critical condenser temperature (T in K; P in bar abs)

$$T^{\text{CRIT}} = \frac{5235}{16.36 + \ln(\alpha_{\text{TOT}}) - \ln(\beta \cdot P)} \quad (11)$$

3. Results and discussion

3.1. Methane

In this section we will, first of all, compare our model with the analysis of Ahmed et al. [1] for methane ($\beta = 4$, $\gamma = 0$), $\text{RH} = 0$ and $P = 1$ bar (ambient pressure, see their Table 1), resulting, as already discussed, in $\alpha_{\text{TOT}} = 3.53$. Using these data we find that the critical temperature in the condenser (at which no net water is produced, $\text{ELW}^* = 0$) is $T^{\text{CRIT}} = 49.4$ °C according to Eq. (10), and $T^{\text{CRIT}} = 49.3$ °C according to Eq. (11). This shows that the approximate expression, Eq. (11), can be safely applied.

Ahmed et al. [1] find that at a condenser temperature of 46 °C the system is a net water producer, with +0.25 mol water produced per mol methane ($\text{ELW}^* = 0.25$). In Fig. 2, calculation results are presented using Eq. (5), which show that at 46 °C + 0.30 mol water/mol fuel is produced according to our analysis, which is very close to the result by Ahmed et al. [1].

Table 1
Comparison of ethanol with ethylene glycol (data for α_{REF} and ζ_1/ζ_2 from [1])

	Ethanol, C ₂ H ₆ O	Ethylene glycol, C ₂ H ₆ O ₂
β	3	3
γ	0.5	1
α_{REF}	0.359	0.293
ζ_1/ζ_2	2	2
Y	2.28	1.92
α_{TOT}	2.64	2.21
$\alpha_{\text{TOT}}/\chi + 1 - \beta/4 + \gamma/2$	13.1	11.3
$(\beta/2)/Y$	0.66	0.78

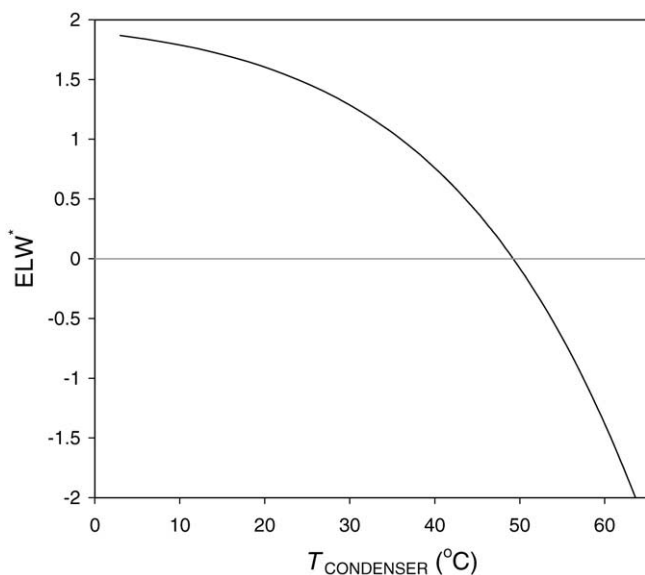


Fig. 2. Excess liquid water produced (moles of water produced per mole of fuel) in methane reforming according to data in Ahmed et al. [1], Table 1 (RH = 0, $P = 1$ bar, $\alpha_{\text{TOT}} = 3.53$, $\beta = 4$, $\gamma = 0$).

3.2. Hydrocarbon fuels

Fig. 3 shows for a range of values for α_{TOT} the critical temperature in the condenser according to Eq. (10) as function of the (absolute) pressure in the condenser P_C and the hydrocarbon fuel composition (β is the ratio of H to C atoms in the fuel, ~ 4 for natural gas, ~ 2 for most liquid fuels). Clearly, the critical temperature in the condenser decreases with increasing α_{TOT} and with decreasing β and P_C . As an example, for $\alpha_{\text{TOT}} = 3.5$, $\beta = 2$ and $P = 1$ bar (absolute) the temperature in the condenser must be below 36°C , which would make operation quite complicated unless ambient temperatures are below 26°C (assuming a 10°C approach temperature, see further on). For hydrocarbon fuels of higher H/C ratio, β , or for higher operating pressures, P_C , water management seems less crucial, even in this most stringent case of RH = 0.

For a relative humidity higher than zero, the calculation of the critical condenser temperature should include the ambient

temperature as well (because it influences p_A^{sat}). According to Ahmed et al. [1] the approach temperature ($T_C - T_A$) is at least $\sim 10^\circ\text{C}$ or else the condenser will become too big. Therefore, we use in Fig. 4 the additional constraint that the ambient temperature, T_A , is 10°C below the critical temperature, $T_C = T^{\text{CRIT}}$, and solve Eq. (10) iteratively. The results are that with increasing humidity the critical condenser temperature rapidly increases, certainly for $P_C = 2$ bar. Indeed, for $P_C = 2$ bar and RH = 60% the critical temperature has disappeared (for the range of α_{TOT} and β presented), and operation is possible under all circumstances.

3.3. Oxygenates

As mentioned, oxygenate fuels of increased oxidation degree, γ , do not produce more water per mole of C-atoms but can be advantageous for water management because of a reduced hydrogen yield, Y , and cathode air flow. As an example we compare ethanol with ethylene glycol using data from Ahmed et al. [1]. Table 1 summarizes the relevant parameters using Eqs. (8) and (9) and shows that an increased γ results in a lower hydrogen yield, Y , and lower overall air-to-fuel ratio α_{TOT} . Consequently, the gas flow to the condenser decreases (term within brackets in Eq. (5)), and last but one entry in Table 1), and the excess liquid water ELW* will increase. Thus, fuels of higher γ require less air and are therefore advantageous from a water management perspective. Table 1 also shows that though the amount of water produced relative to the C-atom flow, $\beta/2$, is independent of oxidation degree, γ , the amount of water produced relative to the hydrogen flow, $\beta/(2 \cdot Y)$ (and thus relative to the electricity produced) increases with increasing oxidation degree, γ . (More water is produced per kWh of electricity with increasing oxidation degree of the fuel.)

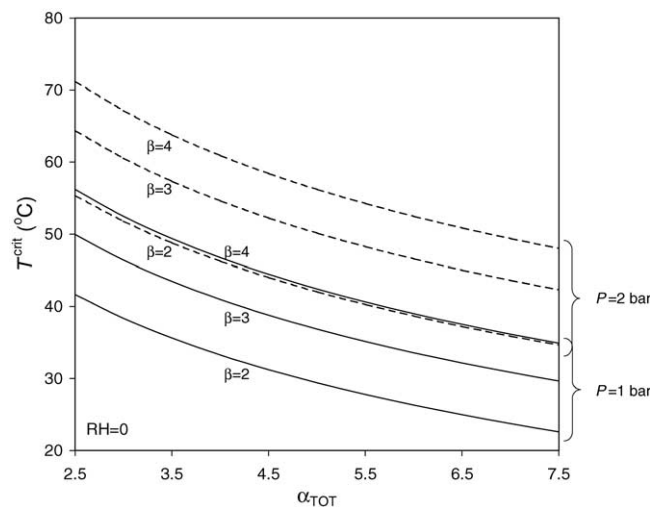
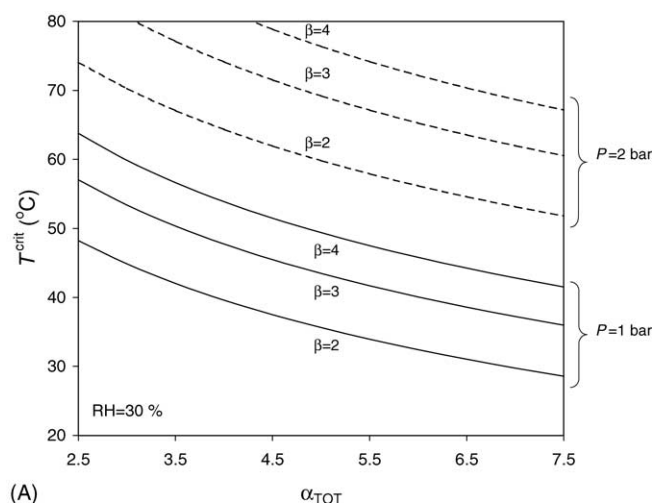
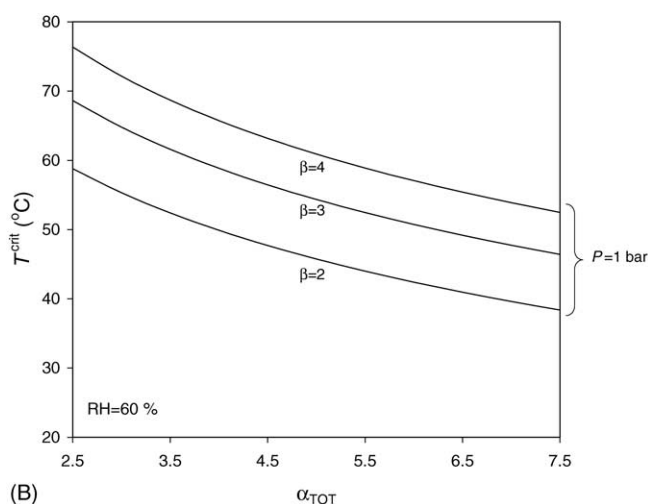


Fig. 3. Critical condenser temperature for water-balanced operation as function of air-to-fuel ratio α_{TOT} , hydrocarbon fuel H/C ratio β and condenser pressure P_C ($\gamma = 0$, RH = 0).



(A)



(B)

Fig. 4. Critical condenser temperature, T^{CRIT} , for hydrocarbon fuels for relative humidities of 30 and 60%. Ambient temperature = $T^{\text{CRIT}} - 10^\circ\text{C}$.

4. Conclusion

A simple mathematical method was presented to calculate the net amount of water produced in a fuel processor fuel cell system that operates on hydrocarbon or oxygenate fuels and uses air as oxidant. By assuming that the net water production is zero, we have calculated the critical condition for water-balanced operation. The model assumes that the amount of hydrogen and hydrocarbons in the exhaust gas (when it is being condensed) is negligible as well as that thermodynamic equilibrium is attained in the condenser.

The relevant parameters are the condenser pressure and temperature, the fuel composition, the overall air-to-fuel ratio and the ambient temperature, pressure and humidity. System parameters such as the steam-to-carbon ratio in the reformer and fuel cell type, pressure and temperature do not directly influence overall water management, but only indirectly via the aforementioned primary parameters such as the overall air-to-fuel ratio.

Typical results are that higher pressures are advantageous as well as a lower air-to-fuel ratio and higher H/C-ratio's of the fuel. For liquid fuels ($\text{H/C} \sim 2$) operating close to atmospheric pressures in moderately hot and dry weather, the methodology suggests that operation can be problematic.

Fuels of higher oxidation degree produce more water per mole of produced hydrogen (thus per kWh electricity), but not per mole of fuel carbon. Because the hydrogen yield and (cathode) air flow are lower, fuels of higher oxidation degree can be advantageous for water-balanced operation.

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