



## Original Research Article

## Influence of anodic gas recirculation on solid oxide fuel cells in a micro combined heat and power system



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## ABSTRACT

The recycle of anode depleted gas has been employed in solid oxide fuel cell systems for the advantage of reusing a fraction of the exhaust rich in steam for the reforming reaction in the cell and reformer. In this work the effect of anode gas recirculation is analyzed at cell and reformer level. The use of a one dimensional fuel cell model gives a novel point of view at cell level of the temperature and gas composition profile stream-wise the cell channel. A baseline configuration without the use of anode off-gas recycling is compared with a configuration which includes the recirculation of the anode gases. Methane is considered as a fuel.

The use of anode gas recycle advantageously increases the cell CO<sub>2</sub> molar fraction in both the reformer and in the cell flow passages. The configuration with the recycle produces higher peak cell temperatures due to a less effective endothermic reforming process; also, the reversed water gas shift reaction is found to increase in CO molar fraction. This produces a lower steam-to-carbon ratio within the cell. In order to mitigate steep temperature gradients, it is found that fuel utilization in the cell-stack should be reduced.

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## Introduction

Solid-oxide fuel cells systems are ideally suited to operate on hydrocarbons, directly converting fuel to electricity. Fuel flexibility is primarily the result of high fuel cell operating temperatures – around 800–1000 °C – which enables many potential applications for SOFCs, including co-generation for stationary applications. The most interesting fuel for SOFC systems used in these applications is natural gas consisting mainly of methane. The direct internal reforming of natural gas in an SOFC is, in principle, the most efficient approach of conversion as it avoids intermediary steps of fuel reforming with their associated thermal losses. However, due to technical limitation such as carbon formation and cell thermal stresses, a pre-reforming process is usually incorporated into the energy system. Steam methane reforming is a viable and the most common pre-reforming process used in these applications. In this case, an external source of steam for the reforming is required. Recycling the anode depleted gas has the advantage to provide steam for the process with the consequence of limiting or even eliminating the use of an external steam production. Development has been focusing on anode off-gas re-use mainly for its advantage to increase the SOFC system efficiency [1].

The aim of the present study is to investigate the effect of the anode off-gas recycling on the fuel cell. This specific area of research has partially been covered in previous analyses. Riensche et al. [2], conducted a system level analysis on a SOFC-based combined heat and power plant (CHP) and found out that the main advantages of anode off-gas recirculation are (i) the elimination of external steam production, (ii) the reduction of cells number in the stack due to lower in-cell fuel utilization, and (iii) a lower steam concentration in the exhaust gas which improves the system thermal efficiency. The electrical efficiency will also increase due to a better heat management. In [1], it is considered that an additional advantage is the reduction of fuel preheating heat transfer by direct contact of fresh fuel and exhaust gas. It is also important to note that the anode off-gas recycle accomplishes the purpose of conserving the de-mineralized and de-ionized water within the system [1].

Many studies on micro combined heat and power systems provided with the recycle of anode gases were carried out. Recent plant-level modeling studies, e.g. [3–7], have analyzed the effect of anode gas recycle on system performance considering parameters such as extent of pre-reforming, air excess ratio etc. Other studies, such as [8,9], have analyzed system performance for hybrid cycle SOFC-Gas Turbine. Peters et al. [10] showed that the increased concentration of CO<sub>2</sub> in the fuel due to anode recycle led to reduced catalytic activity. It was also suggested that recycling is useful to recover waste heat from the anode exhaust

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Abbreviation	
A	area
AGR	anode off gas recycle ratio
CHP	combined heat and power system
CM	carbon methanization
$\dot{E}$	rate of energy flow, (kJ/s)
$\dot{E}_{\text{air}}$	rate of energy in air channel (cathode channel), (kJ/s)
$\dot{E}_{\text{cell}}$	rate of energy in cell structure (anode-electrolyte-cathode-interconnect), (kJ/s)
$\dot{E}_{\text{fuel}}$	rate of energy in the fuel channel (anode channel), (kJ/s)
$\dot{E}_{\text{react}}; \dot{E}_{\text{prod}}$	rate of energy flow in reactant and product, (kJ/s)
$E_N$	Nernst potential, (V)
F	Faraday constant, (A-sec/mol)
$\Delta G$	Gibbs free energy change of the overall reaction, (kJ/kmol)
I	current, (Amp)
K	equilibrium constant, (–)
LHV	lower heating value, (–)
$\dot{m}$	mass flow rate, (g/s)
$n_{0,i}$	initial number of mole of the <i>i</i> th species
$n_e$	number of electrons transferred in the electrochemical reaction
Ni-YSZ	Nickel-Yttria-stabilized zirconia
OCV	open circuit voltage, (V)
<i>p</i>	species partial pressure, (Bar)
$\dot{Q}_{\text{CV}}$	rate of thermal energy transferred across the control volume, (kW)
$\dot{q}$	heat flow rate, (kW)
$\dot{r}_{\text{ox}}$	reaction rate
$R_{\text{cell}}$	total cell resistance, ( $\Omega$ )
$R_U$	universal gas constant, (J/K-mol)
SMR	steam reforming
SOFC	solid oxide fuel cell
STCR	steam to carbon ratio
<i>T</i>	temperature, ( $^{\circ}\text{C}$ )
$U_f$	fuel utilization, (–)
$V_{\text{cell}}$	cell voltage, (Volts)
$W_{\text{CV}}$	rate of work transferred across the control volume, (kW)
WGS	water-gas shift
<i>y</i>	molar fraction, (–)
<i>Greek</i>	
$\xi$	extent of reaction, (–)
$\nu_{ij}$	stoichiometric coefficient of the <i>i</i> th species in <i>i</i> th reaction (–)
<i>Subscript</i>	
act	activation loss
conc	concentration loss
<i>i</i>	species
ohm	ohmic loss
<i>j</i>	reaction
p	primary
Req	requested
Ref	reformer
<i>Subscript</i>	
ox	oxidation
s	secondary; solid

and for the pre-reforming. The amount of recycling was dictated by the amount of steam needed for the pre-reforming process. In an experimental work, Colpan et al. [11] found a negative effect of the fuel recycling on the cell voltage due fuel enrichment in gases with gas species which do not participate in the cell electrochemical reaction.

Furthermore, Evely and Daoudi [12] developed a computational fluid dynamics model to evaluate the potential effectiveness of fuel recycling in mitigating carbon deposits in an internal reforming SOFC. However, this work focused on the window of S:Cs between 0.5 and 1. The test configuration was based on that previously developed by Klein et al. [13] The thermodynamic tendency for carbon deposition through the Boudouard and cracking reactions, two major pathways of carbon deposition at high SOFC operating temperatures, was evaluated within the SOFC anode as a function of fuel humidification and fuel recycling ratio.

Few numerical studies have focused on fuel recycling at cell level. In particular in [14] developed a detailed one dimensional dynamic model of a anode-supported solid oxide fuel cell for both co-flow and counter-flow operation. However, the anode off gas recycle was not considered in this study. In Braun's PhD thesis [15], an SOFC model was developed; however also in this case cell performance were not analyzed in the case of anode gas recycle. In the present work the one-dimensional cell model developed in [15] is employed in order to better analyze the influence of anode off-gas on the fuel cell performance. Comparing fuel cell performance using a 1-D model gives a novel point of view. This offer a better understanding compared to the numerical studies previously mentioned where only zero dimensional SOFC models were employed. Differently from zero dimensional models, a one dimensional model makes possible to analyze thermodynamic properties which would be only shown in a lumped fashion.

Based on the limitations in the reviewed literature, the study motivation is to give a novel point of view at cell level of the temperature and gas composition profiles stream-wise the cell channel and compare the performance in the cases recirculation is applied or not.

## Methodology

To understand and interpret the effect of the recirculation of anode off-gas, two system configurations are defined and compared, (i) a baseline configuration and (ii) a configuration provided with anode gas recycle. Two major component models were considered for the analysis, namely an external-reformer and a SOFC unit. The analysis disregards the other components included in a SOFC system (e.g. Burner, heat exchanger network) as this would not add relevant information to the scope of this study.

The SOFC model provides the fundamental characteristics of solid-cell, anode and cathode gases in the channel direction, whereas the reformer model considers the gas composition at chemical equilibrium. The cell configuration under study is a planar design with air and fuel gas streams in a counter-flow configuration. Fuel utilization is defined as the ratio between moles of hydrogen consumed and hydrogen supplied.

Some general assumptions are made throughout this study:

- *Steady state operation:* SOFC systems will likely be adopted in application operating at steady state conditions (e.g. Combined Heat and Power systems).
- *All gaseous phases are ideal gas:* a gas behaves like as an ideal gas at the temperature and pressure of operation of a SOFC.
- *The components are considered to be adiabatic:* This implies that heat losses are neglected.

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