Journal of Power Sources 313 (2016) 51-64

Contents lists available at ScienceDirect

Journal of Power Sources

journal homepage: www.elsevier.com/locate/jpowsour

Modeling of indirect carbon fuel cell systems with steam and dry gasification



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HIGHLIGHTS

• An equilibrium gasifier model is coupled to a detailed 1-D MEA model.

• Cell performance is greatly enhanced by sending H₂O to the gasifier instead of CO₂.

• Thermal coupling between the gasifier and fuel cell significantly raises efficiency.

• State-of-the-art efficiency is predicted for an indirect carbon fuel cell system.

ARTICLE INFO

Article history: Received 4 October 2015 Received in revised form 1 February 2016 Accepted 15 February 2016 Available online 1 March 2016

Keywords: Carbon fuel cell System efficiency analysis Steam vs. dry gasification Solid oxide fuel cell Exergy analysis

ABSTRACT

An indirect carbon fuel cell (ICFC) system that couples coal gasification to a solid oxide fuel cell (SOFC) is a promising candidate for high efficiency stationary power. This study couples an equilibrium gasifier model to a detailed 1D MEA model to study the theoretical performance of an ICFC system run on steam or carbon dioxide. Results show that the fuel cell in the ICFC system is capable of power densities greater than 1.0 W cm⁻² with H₂O recycle, and power densities ranging from 0.2 to 0.4 W cm⁻² with CO₂ recycle. This result indicates that the ICFC system performs better with steam than with CO₂ gasification as a result of the faster electro-oxidation kinetics of H₂ relative to CO. The ICFC system is then shown to reach higher current densities and efficiencies than a thermally decoupled gasifier + fuel cell (G + FC) system because it does not include combustion losses associated with autothermal gasification. 55–60% efficiency is predicted for the ICFC system coupled to a bottoming cycle, making this technology competitive with other state-of-the-art stationary power candidates.

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1. Introduction

Growing concern over greenhouse gas emissions has driven research into cleaner power production alternatives. However, even with progressive energy policies in place, the IEA projects a global rise in coal-fired power generation over the next few decades due to the industrialization of developing nations [1]. Clean coal technologies with higher system efficiency and CO₂ capture are therefore necessary in the coming decades to mitigate harmful emissions. The carbon fuel cell (CFC) system is a promising candidate for highly-efficient coal power production that releases a diluent-free CO₂ stream ready for capture and sequestration [2]. This paper focuses on a non-molten CFC system design that couples a ceramic solid oxide fuel cell (SOFC) to a gasifier [3].

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http://dx.doi.org/10.1016/j.jpowsour.2016.02.050 0378-7753/© 2016 Elsevier B.V. All rights reserved. Several bench-scale experiments and models have demonstrated the concept of this CFC system with CO₂ as the gasification agent [4–6]. On the other hand, steam gasification has been largely neglected in previous work because it requires more pre-heating and it produces H₂S, which quickly degrades SOFC performance [7]. However, steam has a couple of significant advantages over CO₂ in a CFC system: 1) H₂O gasification proceeds 2–6 times faster than CO₂ gasification [8,9]; and 2) the H₂ produced by steam gasification can be oxidized 2–3 times faster than CO is oxidized on the SOFC anode [10]. Therefore, there is a strong incentive to consider H₂O as an alternative to CO₂ in a CFC system to improve performance.

Two fundamental questions still need to be answered for this CFC system: 1) how much will system power density increase if steam is used instead of carbon dioxide as the gasifying agent?; and 2) how much will system efficiency increase when a gasifier is coupled to a SOFC? A preliminary CFC modeling study on this topic coupled an equilibrium gasifier to a SOFC with fixed voltage and





Nomenclature

A_s	specific area of nickel per cell footprint area
e _i	total exergy of species i (J mol ⁻¹)
e_i^{cn}	chemical exergy of species i (J mol ⁻¹)
\dot{E}_{G}^{Q}	rate of exergy transferred by heat to gasifier (W cm ⁻²)
E	potential difference across anode/electrolyte double-
F	equilibrium potential difference across anode/
Leq	electrolyte double-laver (V)
Ecell	operating cell voltage (V)
Erev	reversible cell voltage (V)
F	Faraday constant ($C \text{ mol}^{-1}$)
ΔG_i	Gibbs free energy change of reaction i (J mol ⁻¹)
i	total cell current density (A cm^{-2})
i _{co}	current density produced by CO electro-oxidation
	$(A \text{ cm}^{-2})$
i_{H_2}	current density produced by H_2 electro-oxidation
	$(A \text{ cm}^{-2})$
ι ₀	exchange current density (A cm^{-2})
ι_{H_2}	exchange current pre-factor (A cm ⁻² Pa ⁻¹)
Ji,m	molar flux of species <i>i</i> at node $m \pmod{s^2}$
К _f ,Кb	(mol s ⁻¹ m ⁻¹)
K _{eq,i}	chemical equilibrium constant of reaction <i>i</i>
Kg	total number of gaseous species in the anode
l _{TPB}	triple-phase-boundary length (m cm ⁻²)
L _{el}	electrolyte thickness (cm)
n _i	molar flow-rate of species $i \pmod{s^{-1} \text{ cm}^{-2}}$
p_i	partial pressure of gas species <i>i</i> (Pa)
P	total pressure (Pa)
Q_{FC}	rate of heat release from fuel cell ($W \text{ cm}^{-2}$)
Q _G	rate of heat input to gasifier (W cm^{-2})
Q _{rej}	net rate of heat rejected by system (W cm^{-2})
R	universal gas constant (J mol $^{-1}$ K $^{-1}$)

current [11]. That study compared a few CFC systems with varying degrees of coupling between components, and used thermodynamic analyses to compare the performance of these systems with both CO₂ and H₂O. Although the paper gave a good overview of the different systems, it did not find an advantage of using H₂O over CO₂ because the model fixed cell voltage and did not include finiterate kinetics for SOFC surface reforming and electrochemistry. Similarly, that thermodynamic analysis did not accurately assess the efficiency gained by thermal coupling because a finite-rate kinetic model is needed to accurately determine fuel cell efficiency.

This paper addresses these fundamental questions about the CFC system using an equilibrium gasifier model coupled to a 1D-MEA model with finite-rate kinetics. The gasifier can be modeled in equilibrium because it is in a separate compartment upstream of the SOFC stack and can therefore be sized to ensure sufficient residence time. The SOFC model, on the other hand, includes finite-rate kinetics in order to accurately compare the H₂ and CO electrochemical oxidation rates and assess the various losses in the cell. First the CFC system model is presented along with an explanation of the different CFC system classifications. Next the governing equations are presented for the gasifier and fuel cell components of the model. Finally, the two fundamental questions posed earlier are addressed sequentially with two modeling studies. The first study compares the impacts of H₂O and CO₂ gasification on system

R_{el}	ionic resistance in the electrolyte ($\Omega \text{ cm}^2$)
\$surf.k	rate of production of gas species k by surface reactions
5	$(mol s^{-1} cm^{-2})$
T_i	temperature of component i (K)
u _f	fuel utilization
u_{O_2}	oxygen utilization
Xi	mole fraction of gas species <i>i</i>
Ŵ	electrical power output (W cm ⁻²)
Greek le	etters
η _i	efficiency of component or system <i>i</i>
$\eta_{act,a}$	anode activation overpotential (V)
$\eta_{act,c}$	cathode activation overpotential (V)
$\eta_{con,a}$	anode concentration overpotential (V)
$\eta_{con,c}$	cathode concentration overpotential (V)
η_{ohm}	ohmic overpotential (V)
σ_0	ionic conductivity of electrolyte (S cm^{-1})
$\theta_{O^-,YSZ}$	surface coverage of O ⁻ ions on YSZ
θ_{YSZ}	vacancy coverage of YSZ
Supersc	ripts
ex	exergy
Subscrij	ots
а	anode
An	anode exhaust stream
С	cathode
Са	cathode exhaust stream
G	gasifier
FC	fuel cell
R	recycle stream
sys	system without bottoming cycle
sys, bot	tom system with bottoming cycle
ТРВ	triple-phase-boundary

performance over a range of temperatures. The second study explores the impact of thermal coupling between the gasifier and fuel cell on system efficiency.

2. Model description

The objective of this model is to determine CFC system power and efficiency over a range of current densities and operating conditions. The combined CFC system is presented first, followed by descriptions of the equilibrium gasifier model and fuel cell model. The fuel cell model has been previously validated against experimental data for many syngas mixtures at 800 °C, and the governing equations of the model are described in detail elsewhere [12,13].

2.1. The CFC system model

2.1.1. System classifications

There is considerable debate over the proper terminology to describe CFC systems [14]. Table 1 breaks down the CFC system classification adopted here by the extent of coupling between the gasifier and SOFC. Systems listed higher are more coupled and theoretically capable of obtaining higher efficiencies, but at the expense of more complex integrated designs. The model in this paper focuses on the indirect CFC (ICFC) and gasifier + fuel cell

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