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A transient analysis of three-dimensional heat and mass transfer in a molten carbonate fuel cell at start-up

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ABSTRACT

In this paper, we investigate the time-dependent heat and mass transfer in a molten carbonate fuel cell at start-up. Thus, a three-dimensional, transient mathematical model is presented through a comprehensive inclusion of various physical, chemical and electrochemical processes that occur within the different components of molten carbonate fuel cells. The model is proposed as a predictive tool to provide a three-dimensional demonstration of variable variations at system start-up. The local distribution of field variables and quantities are showcased. It reveals that the electrochemical reaction rate is dominated by the over-potential, not by the reactants' molar fraction. Reversible heat generation and consumption mechanisms of the cathode and anode are dominant in the first 10 s while the heat conduction from the solid materials to the gas phase is negligible. Meanwhile, activation and ohmic heating have nearly the same impact within the anode and cathode. Based on these findings, the importance of heat conduction and its main features are finally assessed.

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Introduction

The important advantages of molten carbonate fuel cells (MCFCs) such as low emissions, flexibility in fuel choice and high energy conversion efficiency, have initiated research and development of MCFCs for applications in stationary and mobile power generation units. Despite technical advances, capital costs, cell corrosion and degradation still remain large obstacles to commercialization. Hence, the main aim of the ongoing MCFC studies is to overcome these challenges. Therefore, it is crucial to extend cell life with a stable

performance [1] and make the system more efficient so as to reduce operating and maintenance costs.

Technically, the non-uniformity of temperature and current density profiles, caused by poor conductivities, can accelerate cell degradation and reduce the effective lifetime [2]. In view of this, an improved understanding of the underlying transport and electrochemical phenomena is essential. By reviewing previous efforts in the literature, it is readily apparent that modeling and numerical studies have played a key role in providing better insight into the complex processes occurring in stack and particular unit cell components [3].

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Nomenclature			
A_v	electrode active surface area, $\text{m}^2 \text{m}^{-3}$	θ	electrolyte filling degree
c_p	specific heat, $\text{J kg}^{-1} \text{K}^{-1}$	η	over-potential, V
D	mass diffusivity, $\text{m}^2 \text{s}^{-1}$	μ	dynamic viscosity, $\text{kg m}^{-1} \text{s}^{-1}$
E_{eq}	equilibrium electric potential, V	ν	species stoichiometric coefficient of the reaction
F	Faraday's constant, $96,485, \text{C mol}^{-1}$	ρ	density, kg m^{-3}
f	frequency, Hz	ϕ	electric potential, V
i_0	exchange current density, A m^{-2}	σ	electric conductivity of the solid phase, S m^{-1}
i_0^0	reference exchange current density, A m^{-2}	κ	electric conductivity of the electrolyte phase, S m^{-1}
J	local current density, A m^{-2}	<i>Subscripts and superscripts</i>	
R	volumetric current density, A m^{-3}	a	anode
k	thermal conductivity, $\text{W m}^{-1} \text{K}^{-1}$	agc	anode gas channel
K	permeability, m^2	c	cathode
M	molecular weight, g mol^{-1}	cgc	cathode gas channel
n	number of electrons	e	electrolyte phase
P	static pressure, Pa	g	gas phase
R	universal gas constant, $8.314 \text{ J mol}^{-1} \text{K}^{-1}$	in	inlet
S	molar entropy, $\text{J mol}^{-1} \text{K}^{-1}$	i	ith component
S	source terms	j	jth species
t	time, s	m	mass equation
T	temperature, K	out	outlet
\vec{v}	gas velocity, m s^{-1}	s	solid phase
X	molar fraction	T	energy equation
Y	mass fraction	u	momentum equation
<i>Greek letters</i>		ϕ_e	electronic charge equation
α	transfer coefficient	ϕ_c	carbonate ion charge equation
τ	tortuosity	eff	effective
ϵ	porosity	ref	reference state

In the 1980s and 1990s, MCFC modeling was mostly focussed on electrode-level models [4–12]. The contributions of some key models and their associated weaknesses and advantages are summarized in Table 1. The studies that have implemented electrode-level models mostly illustrate the transport phenomena that take place in just one or two components of the unit cell and hence simplify the effects of the adjacent components, using a fixed boundary condition. Nevertheless, over the past decade researchers have employed electrode-level models and have modified them to investigate different aspects of unit cells.

However, advancing the mathematical models from electrode-level to cell-level has not always incorporated all physically relevant phenomena, conditions and characteristics due to various simplifying assumptions. In other words, model reduction methods are widely used to establish a simple simulation model at various operating conditions [2].

For instance, Lukas et al. [13] employed the lumped-parameter formulation of the first principle equations with a simplifying assumption indicating that the solid mass temperature is equal to the exit stream temperature. A model developed by Fermegila et al. [14] neglected the enthalpy transport between the electrode pores and gas phase, and also assumed an equally distributed current pattern in both electrodes. Hao et al. [15] developed a model assuming identical heat characteristics for anode, cathode and electrolyte while they typically have an order of magnitude difference.

Heidebrecht and Sundmacher [16,17] have also developed a transient model which is based on constant pressure in the gas phase along with a lumped solid phase for energy balance. Lee et al. [18] presented a mathematical model to simulate the performance of a molten carbonate fuel cell 5 kW stack. They assumed identical average current densities for each cell of the stack. A three-dimensional model was also developed by He and Chen [19]. Technically, their model can consider simultaneously the dominant processes such as mass transport, chemical reactions and heat transfer. It is also capable of calculating the local mass concentration but still uses an empirical correlation for the cell current densities instead of solving the fundamental conservation equation. The authors later showed [20] how the current density profile changes rapidly in the beginning and slowly in the following stage, and that the temperature response is slow.

It is apparent that a considerable number of the available studies have been carried out by either performing the simulation for individual MCFC components or reducing the dimension to 0D, 1D or 2D. In addition, many studies have employed uniform distribution for some of the field variables instead of finding their values by solving the corresponding PDEs, which significantly influences the accuracy of the dynamic results. In other words, the above mentioned assumptions that are implemented in the previous studies, reduce the accuracy of the results and hence the reliabilities of the model for further steps, e.g., control system design.

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