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Analysis of cold start processes in proton exchange membrane fuel cell stacks

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HIGHLIGHTS

- ► A 3D multiphase model is developed to study the cold start process of PEMFC stacks.
- ► Stacks with more cells can reach higher temperature with better performance.
- ► Ice formation in middle cells is slower.

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ABSTRACT

To comprehensively understand the cold start processes of proton exchange membrane fuel cell (PEMFC) stack which is important for the automotive applications, a three-dimensional multiphase PEMFC stack model is developed in this study. The detailed analysis of the cold start processes shows that for the stacks with more cells, the voltage decreases more slowly due to the lower ice formation rates. The temperature increases faster for a stack with more cells, and a higher temperature can be reached at the end of the cold start process. No apparent difference in voltage exists among the different individual cells in a stack when the reactant gases are evenly supplied to each cell. The temperature in the individual cell in the middle of a stack is higher and more evenly distributed than those on the sides and single cells, due to weakened cooling effect of the bi-polar plate (BP) on the membrane electrode assembly (MEA), and the ice formation rate is also lower in the middle cell. At a lower current density, the ice in the cathode catalyst layer (CL) is formed faster at the section close to the BP, and it is close to the membrane at a higher current density.

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1. Introduction

Proton exchange membrane fuel cell (PEMFC) is one of the promising clean and efficient power sources for transportation applications, owing to its high power density, low temperature operation and zero/low emission [1]. Start-up of PEMFC stack from sub-zero temperatures has been recognized to be an essential issue before its successful commercialization. Several countries and regions have set specific targets for cold start performance. For example, in the United States, the latest target was set by the Department of Energy (DOE), which requires unassisted successful start-up from −40 °C [2], a much more difficult requirement than the one established in 2005. The European Union sets several general technical targets for the years 2015–2020, including the lowest successful cold start temperature of −25 °C, and well maintained proton conductivity at low temperatures (over 10 mS cm^{-1} at −20 °C) [3]. The Japanese government has also established a series of programs, and in recent years, the main plan has been shifted from “strategic development” to “commercialization promotion” [4], indicating that the focus of research and development is now on the commercially applied fuel cell stacks. As for fuel cell manufacturers, the General Motors Corporation set the target of unassisted start-up from −40 °C as well [5].

A number of experimental studies have been conducted to investigate the cold start performance and characteristics of PEMFCs [6–18]. The focus has been on the measurement of general performance behaviors, performance degradation and visualization of ice formation during cold start processes. Cold start models have also been developed for single cells [19–24] and stacks [25–27] in an effort to better understand the start-up process. Single cell analytical models have been mainly targeted to discover the relationship between the design/operating parameters and cold start performance [19–21], and most of these models have only considered either individual components (e.g. cathode catalyst layer (CL)) [19] or simplified cells (one-dimensional models) [20,21]. Multiphase multi-dimensional numerical simulations have been carried out for single PEMFCs to investigate more detailed transport processes [22–24]. Mao et al. [22] found that ice in the cathode CL appears first at the flow channel inlet region and extends

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Nomenclature

A	stack geometric area, m^2
c	molar concentration, mol m^{-3}
C_p	specific heat, J kg^{-1} K^{-1}
D	mass diffusivity, m^2 s^{-1}
EW	equivalent weight of membrane, 1100 kg kmol^{-1}
F	Faraday's constant, 96,487 C mol^{-1}
h	latent heat, J kg^{-1}; heat transfer coefficient, W m^{-2} K^{-1}
I	current density, A cm^{-2}
j	reaction rate, A m^{-3}
k	thermal conductivity, W m^{-1} K^{-1}
K	permeability, m^2
$\dot{m}$	mass flow/transfer rate, kg s^{-1}
M	molecular weight, kg kmol^{-1}
p	pressure, Pa
q	heat flux, W m^{-2}
$\dot{Q}$	heat transfer rate, W
RH	relative humidity
s	volume fraction
S	source terms, entropy, J kmol^{-1} K^{-1}
t	time, s
T	temperature, K or °C
$\vec{u}$	velocity, m s^{-1}
x	mole fraction
Y	mass fraction

Greek letters

α	transfer coefficient
ε	porosity
η	overpotential, V
ι	interfacial drag coefficient
κ	electrical conductivity, S m^{-1}
λ	water content in ionomer
μ	dynamic viscosity, kg m^{-1} s^{-1}
ξ	stoichiometry ratio
ρ	density, kg m^{-3}
ϕ	electrical potential, V
ω	volume fraction of ionomer in catalyst layer

Subscripts and superscripts

a	anode
act	activation
BP	bi-polar plate
c	cathode
CL	catalyst layer
cond	condensation
desb	desublimation
eff	effective
ele	electronic
equil	equilibrium
EOD	electro-osmotic drag
evp	evaporation
f	frozen
fl	fluid phase
fmw	frozen membrane water
fusn	fusion
FPD	freezing point depression
g	gas phase
GDL	gas diffusion layer
H_2	hydrogen
H_2O	water
i	the ith components or the ith cell in a stack
ice	ice
in	inlet
init	initial condition
ion	ionic
lq	liquid water
m	mass (for source term)
melt	melt
mem	membrane
N	total number of cells in a stack, normal condition
nf	non-frozen
nmw	non-frozen membrane water
O_2	oxygen
out	outlet
pc	phase change
ref	reference state
sat	saturation
sl	solid phase
stk	stack characteristic
surr	surroundings
T	energy (for source term)
u	momentum (for source term)
vp	water vapor
wall	surrounding wall of the single cell or stacks
0	intrinsic value
l-i	liquid water to ice (vice versa)
n-f	non-frozen membrane water to frozen membrane water (vice versa)
n-i	non-frozen membrane water to ice
n-v	non-frozen membrane water to vapor (vice versa)
v-i	vapor to ice
v-l	vapor to water liquid (vice versa)

toward the outlet region gradually, the start-up current density influences the water uptake potential of the membrane, and a lower current density is beneficial for water uptake. Meng [23] predicted that the cold start process obtains a benefit from the higher gas flow rates in the cathode flow channel. Jiao and Li [24] found that it is favorable to increase the ionomer fraction in the cathode CL, and a thinner membrane enhances the water uptake rate.

In order to supply sufficient power for vehicles, individual PEMFCs are often assembled in series to form stacks. Interactions between the individual cells in a stack become significant, making the cold start characteristics of PEMFC stacks different from single cells [16]. Sundaresan and Moore [25] developed a layered cold start stack model, which considered the water phase change and the thermal effects, and this model predicted the temperature of different cells in a stack. A one-dimensional model developed by Khandelwal et al. [26] also obtained the temperature distribution in a PEMFC stack. Based on this model, several assisted start-up strategies were tested and compared. Ahluwalia and Wang [27] developed a two-dimensional cold start stack model, and the ice formation was considered in this model. It was found that a high current density is favorable for rapid cold start of PEMFC stacks, and the metal bi-polar plates (BPs) are better than graphite to improve the cold start abilities due to the lower heat capacity of metal. The PEMFC cold start related studies were also reviewed recently in detail by Meng and Ruan [28] and Jiao and Li [29].

As mentioned above, most of the previous cold start models only considered single cells and one-dimensional/two-dimensional approach to stack level. To comprehensively understand the PEMFC stack cold start processes, a three-dimensional multiphase PEMFC stack model is needed. In this study, a three-dimensional

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