Journal of Power Sources 258 (2014) 89–97

Contents lists available at ScienceDirect

Journal of Power Sources

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

A three-dimensional pore-scale model of the cathode electrode in polymer-electrolyte membrane fuel cell by lattice Boltzmann method

G.R. Molaeimanesh, M.H. Akbari*

Center for Fuel Cell Research, School of Mechanical Engineering, Shiraz University, Molla-Sadra Ave., Shiraz 71348-51154, Iran

HIGHLIGHTS

- A 3-D lattice Boltzmann model of cathode GDL in PEM fuel cell is presented.
- Non-homogeneity and anisotropy of GDL, and electrochemical reaction are included.
- General orientation of GDL carbon fibers, shapes species and current distributions.
- Normally oriented carbon fibers cause larger current variations on catalyst layer.
- Microstructure must be regarded as an important characteristic of any GDL material.

ARTICLE INFO

Article history:
Received 25 December 2013
Received in revised form
31 January 2014
Accepted 6 February 2014
Available online 18 February 2014

Keywords:
PEM fuel cell
Lattice Boltzmann method
Three-dimensional model
GDL microstructure
Electrochemical reaction

ABSTRACT

High power density, low operation temperature, high efficiency and low emissions have granted proton exchange membrane (PEM) fuel cells the most promising future among all types of fuel cells. The porous electrodes of PEM fuel cells have a complicated, non-homogeneous, anisotropic microstructure. Therefore, pore-scale modeling techniques such as lattice Boltzmann method, which can capture non-homogeneous and anisotropic microstructures, have recently gained a great attention. In the present study, a three-dimensional lattice Boltzmann model of a PEM fuel cell cathode electrode is proposed in which electrochemical reaction on the catalyst layer and microstructure of GDL are taken into account. The model enables us to simulate single-phase, multi-species reactive flow in a heterogeneous, anisotropic gas diffusion layer through an active approach. To show the capability of the proposed model, reactive flow in three reconstructed GDLs with different anisotropic characteristics is simulated to investigate the effects of GDL microstructure on species and current density distributions. The results demonstrate that when carbon fibers are more likely oriented normal to the catalyst layer, species density distribution is thicker and more disturbed. Current density also experiences a larger variation on the catalyst layer in such a case.

1. Introduction

A sustainable, high quality life is the basic driver for providing a clean, safe, reliable and secure energy supply in the world. Fuel cells and hydrogen systems are important technologies which may contribute positively to the future world energy demand. Among different types of fuel cells, proton exchange membrane (PEM) fuel cells currently hold the most promising prospect. They can be employed in a wide range of applications, ranging from very small portable devices such as mobile phones and laptops, through transport applications like cars, delivery vehicles, buses and ships, to combined heat and power generators in stationary applications in the domestic and industrial sectors [1].

Nevertheless, some barriers such as high cost and operational difficulties hinder PEM fuel cells' wide commercialization, which make fundamental research for their development inevitable. Modeling of reactant gas transport accompanied by electrochemical reactions in the electrodes is of great importance, especially in the cathode where the oxygen reduction reaction is sluggish and inefficient [2]. Numerous numerical models of the cathode electrode with different features can be found in the literature [3]. In most of these models, the gas diffusion layer (GDL) of cathode electrode is considered as a homogeneous and isotropic porous medium, while in reality that is not the case [4]. In this regard, pore-scale modeling techniques which can realistically capture the microstructure of the cathode GDL are extremely attractive.

Some researchers have used more conventional methods, such as the finite volume, to model electrodes of a PEMFC. Harvey et al. [5], for example, used such a method to investigate different approaches to

* Corresponding author. Tel.: +98 917 308 8424; fax: +98 711 647 3511.
E-mail addresses: h-akbari@shirazu.ac.ir, hakbari391@yahoo.com (M.H. Akbari).

http://dx.doi.org/10.1016/j.jpowsour.2014.02.027
0378-7753/

Nomenclature

a	roughness factor
$\vec{c}$	particle velocity (lu ts^{-1})
c_s	speed of sound in lattice (lu ts^{-1})
D	diffusivity (m^2 s^{-1})
D^{LB}	lattice Boltzmann diffusivity
Da	Damköhler number
F	Faraday's constant (A s mol^{-1})
Fo	Fourier number
f	density distribution function
j	current density (A m^{-2})
k	rate constant (m s^{-1})
k^{LB}	lattice Boltzmann rate constant
l	characteristic length scale (m)
MW	molar mass (kg kmol^{-1})
P	pressure (Pa)
R_u	universal gas constant (J mol^{-1} K^{-1})
$\vec{r}$	particle position vector (lu)
r''	reaction rate per unit surface area (mol m^{-2} s^{-1})
T	temperature (K)
t	time (ts)
$\vec{u}$	velocity vector (lu ts^{-1})
$\vec{u}'$	composite velocity vector in Eq. (5) (lu ts^{-1})
w	weighting factor

Greek symbols

α	transfer coefficient
η	activation over-potential
λ_n	Eigen-parameter in Eq. (11)
μ	dynamic viscosity (kg m^{-1} s^{-1})
ν	kinematic viscosity (lu^2 ts^{-1})
ρ	density (lm lu^{-3} or kg m^{-3})
ρ^*	dimensionless density
τ	relaxation time (ts)

Subscripts and superscripts

A	type A species
C	type C species
eq	equilibrium
i	direction i of lattice
k	species k
LB	lattice Boltzmann
n	nitrogen
o	oxygen
ref	reference
sr	surface reaction
w	water

modeling the catalyst layer of PEM fuel cells. They compared three catalyst treatments of thin-film model, discrete-catalyst volume model and agglomerate model, and concluded that only the agglomerate model could capture mass transport limitations that occur at high current densities. Sahraoui et al. [6] presented a two-dimensional, finite-volume model of a PEM fuel cell taking into consideration the finite thicknesses of the catalyst layer and membrane. Sun et al. [7] also introduced a two-dimensional model for PEM fuel cell cathode that treated the catalyst layer as agglomerates of polymer electrolyte coated catalyst particles. They showed that the cathode over-potential inside the catalyst layer was non-uniform influenced by the channel-land geometry.

In the past decade, lattice Boltzmann method has emerged as a powerful pore-scale modeling technique applicable in complicated, heterogeneous, anisotropic porous media such as GDLs of PEM fuel cells. Lattice Boltzmann method is superior to other conventional numerical modeling techniques through many aspects such as capability of dealing with complex boundaries of a complicated microstructure, easy parallelizable algorithm development, and facility in modeling multi-phase fluid flow in a porous medium [8].

However, a significant challenge for lattice Boltzmann modeling of the cathode electrode is a proper way to consider electrochemical reaction in the catalyst layer [9]. To the best of the authors' knowledge the electrochemical reaction is taken into account in only a few two-dimensional lattice Boltzmann investigations, such as those presented by Chen et al. [10–12]; moreover, no such three-dimensional modeling, taking into account the electrochemical reaction, was found in the literature. In the present study, a single-phase, three-dimensional model of the cathode electrode of a PEM fuel cell is presented in which the catalyst layer is considered as a thin interface where the electrochemical reaction occurs. The proposed model is implemented to investigate the effects of GDL microstructure on the distributions of species and current density. An advantage of the proposed model over previous lattice Boltzmann models [10–12] is the use of an active approach for species transport. In the preceding investigations [10–12], a passive approach was applied in which the velocity field is only solved for nitrogen that is taken as the solvent. This may lead to lack of accuracy especially when the density of other species is comparable to that of nitrogen which may happen at high over-potentials; in such cases no species can be considered as a solvent [13]. On the other hand, in an active approach the internally coupled velocity fields of all species are solved individually which can lead to more accurate results.

2. Numerical method

2.1. Framework of lattice Boltzmann method

Lattice Boltzmann method is a powerful numerical modeling technique that has attracted much interest in the past decade. In this method, a fluid is assumed to be composed of virtual fluid particles which move and collide with each other in a lattice structure. These particles are treated by a distribution function rather than by positions and velocity vectors [14]. This kinetic nature of lattice Boltzmann method provides some superior features relative to other numerical methods, such as the linearity of the transport equation (in comparison with nonlinear Navier–Stokes equations), calculation of pressure by an equation of state (in comparison with calculating the pressure by solving Poisson differential equation), etc. [8].

The present numerical model is based on the lattice-Boltzmann method with a single relaxation time collision operator (the so-called BGK model [15]) and the popular D3Q19 lattice scheme. The 3 in D3Q19 refers to the number of lattice dimensions and the 19 denotes the number of possible directions for a particle movement in the lattice as shown in Fig. 1.

The lattice Boltzmann equation is derived from simplification of Boltzmann equation in a lattice [16]. It can be expressed as

$$f_i(\vec{r}+\vec{c}_i\Delta t, t+\Delta t) = f_i(\vec{r},t) + \frac{\Delta t}{\tau}\left[f_i^{eq}(\vec{r},t) - f_i(\vec{r},t)\right] \quad (1)$$

where f_i is the density distribution function in direction i, f_i^{eq} is the equilibrium density distribution function, $\vec{r}$ refers to space position, t is time, $\vec{c}_i$[1] is the particle velocity vector in direction i, and τ

[1] lu and ts are the unit of length and time in lattice Boltzmann method, respectively.