



Effective diffusivity in partially-saturated carbon-fiber gas diffusion layers: Effect of through-plane saturation distribution



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ABSTRACT

The effective diffusivity of gaseous species in partially-saturated finite-size porous media is a valuable parameter for mathematical modeling of many processes, but it is difficult to measure experimentally. In this work, the effective diffusivity of carbon-fiber gas diffusion layers (GDLs) used in polymer electrolyte fuel cells (PEFCs) was determined by performing lattice Boltzmann (LB) simulations on X-ray tomographic reconstructions of invading water configurations. Calculations on dry GDLs were in close agreement with previous experimental data; the effective diffusivity was reduced by the addition of PTFE due to the loss of pore volume and the higher tortuosity of transport paths. The effect of water saturation was significantly larger. It was found that the resistance of water to gas transport was extremely dependent on the saturation distribution through the porous medium, particularly the peak saturation, and not just the average saturation as is typically considered in the literature. Through-plane diffusion was dramatically limited in materials with high-peak local saturations, even at low average saturation levels. No significant limitations were observed for diffusion in the material plane. The computed results demonstrate the strong sensitivity of finite-size porous media to local conditions, highlighting the difficulties of applying volume-averaged continuum-scale modeling techniques to micro-scale materials.

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1. Introduction

Polymer electrolyte fuel cells (PEFCs) are a leading candidate to replace internal combustion engines for automotive applications, as they are the only presently available technology that can offer equivalent power density, range and refueling times in a sustainable way. The electrochemical conversion of reactants in PEFCs leads to virtually zero emissions of pollutants (water and heat are the only products generated in the cell), and provides 2–3 times higher efficiency as compared to high-temperature combustion devices [1]. However, engineering factors such as high cost and limited durability remain significant barriers to large-scale commercialization. To reduce cost, it is imperative that the mass of platinum (Pt) catalyst per vehicle be reduced significantly; this can be achieved by lowering the loading of Pt per unit active cell area, or reducing the active area (or both). The latter approach, however, requires an increase of the current density (A/m²) of

the cells, to maintain the output power (W) of the stack. Therefore, thorough understanding of the transport of reactants and products to/from the catalyst sites in PEFCs is needed [2–5].

One key element of a fuel-cell electrode is the gas diffusion layer (GDL), which is a thin highly porous layer composed of carbon fibers. The GDL must fulfill several functions such as providing structural stability to the cell, and transport pathways for gaseous reactants and liquid water through its pore volume and electronic current and heat through its solid structure. A deep knowledge of the effects of liquid water on the gas diffusivity of GDLs is essential to achieving an increased PEFC performance [2–5].

The standard approach for modeling the effective diffusivity in porous materials is to modify the bulk diffusion coefficient, D_{bulk} , to get an effective diffusivity, D^{eff} , through the porous domain [6],

$$D^{\text{eff}} = D_{\text{bulk}} \frac{\varepsilon_V}{\tau} \quad (1)$$

where ε_V is the void volume fraction of the porous medium, and τ is the so-called tortuosity, which accounts for the indirect route that gas molecules must follow to traverse the ‘tortuous’ pore space. In porous media whose pore structure is partially filled with liquid

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Nomenclature

Symbols

A	GDL cross-sectional area [LB units]-[m ²]
C	mass concentration [LB units]-[mol/m ³]
c	lattice velocity [LB units]-[m/s]
c_s	lattice 'speed of sound' [LB units]-[m/s]
D	mass diffusivity [LB units]-[m ² /s]
d	diameter [m]
d_p	pore diameter [m]
f	particle distribution function [LB units]
$f(\varepsilon)$	normalized dry effective diffusivity function; see Eq. (3)
g	gravitational acceleration [m/s ²]
$g(s)$	relative effective diffusivity; see Eq. (3)
h	height [m]
I	grayscale intensity
j	Fickian diffusion flux [LB units]-[mol/m ² s]
L_i	GDL sample length in i -direction [LB units]-[m]
n	best-fit power-law exponent
p	pressure [Pa]
p_c	capillary pressure [Pa]
R	radius [m]
s	liquid saturation
$\bar{s}_x(x_i)$	yz-averaged liquid saturation at slice $x = x_i$
$\bar{s}_z(z_i)$	xy-averaged liquid saturation at slice $z = z_i$
t	time [LB units]-[s]
Δt	time step [LB units]-[s]
V	volume [LB units]-[m ³]
x	in-plane coordinate [LB units]-[m]
x_i	node position [LB units]-[m]
Δx	spatial increment [LB units]-[m]
y	secondary in-plane coordinate [LB units]-[m]
z	through-plane coordinate [LB units]-[m]

Greek letters

α	index
δ_z	thickness window length of the GDL in the LBM simulations [m]; see Fig. 7
ε	porosity
$\bar{\varepsilon}_z(z_i)$	xy-averaged porosity at slice $z = z_i$

ε_V	void volume fraction
$\lambda(s)$	saturation-dependent function; see Fig. 10
ρ	density [kg/m ³]
τ	tortuosity/dimensionless relaxation time
ϕ_V	solid volume fraction
ψ	parameter defined in Eqs. (8) and (9)

Subscripts

avg	volume-averaged quantity in the whole sample
bulk	bulk conditions
CD	computational domain
crit	critical local transport conditions; see Figs. 9 and 12
D/C	dilation/contraction
F	fiber
FT	full thickness
g	gas phase
IP	in-plane direction
in	inlet/inner
j	direction of interest or under study
l	liquid phase
out	outlet
T	total
TP	through-plane direction
th	threshold
w	water

Superscripts

char	characteristic
eff	effective
eq	equilibrium
flat	flat saturation distribution
dry	dry conditions
lin	linear saturation distribution
max	maximum
min	minimum
wet	wet or partially-saturated conditions
*	dimensionless or normalized value

water, gas diffusion is further blocked since liquid offers essentially no gas flux. The configuration, distribution, and extent of water within the pore space have major impacts on the diffusion rates, above and beyond the pore-structure effects [7]. To differentiate the impacts of the pore/throat structure of the dry material from those of the invading liquid, Eq. (1) can be decomposed into two independent functions, one accounting for the effect of the unsaturated medium, $f(\varepsilon)$, and another for the relative effect of liquid in the pore space, $g(s)$:

$$\frac{D^{\text{eff,wet}}}{D_{\text{bulk}}} = \frac{D^{\text{eff,dry}}}{D_{\text{bulk}}} \frac{D^{\text{eff,wet}}}{D^{\text{eff,dry}}} = f(\varepsilon)g(s) \quad (2)$$

where ε is the porosity, s is the liquid saturation (defined as the ratio of liquid volume to pore volume), and $D^{\text{eff,dry}}$ and $D^{\text{eff,wet}}$ are the effective diffusivities of the porous medium under dry, and partially-saturated (or wet) conditions, respectively. A review of previous studies attempting to determine $f(\varepsilon)$ and $g(s)$ for fuel-cell GDL materials is presented below.

Multiple experimental [8–19], numerical [20–22] and semi-analytical [23–28] works on the dry effective diffusivity function, $f(\varepsilon)$, for carbon-paper GDLs can be found in the literature. These investigations provide fundamental information about the effects of compression, the fabrics of the substrate, the addition of

wet-proofing agents, and the incorporation of micro-porous layers (MPLs) on the effective gas diffusivity of GDLs. Good agreement is observed among different authors. All of them show that the effective diffusivity of many carbon-fiber papers is anisotropic, with higher values in the in-plane direction due to preferential alignment of the GDL fiber structure in the material plane [8]. Moreover, it has been generally found that the effective diffusivity is notably lower than predicted by most established and widely used models, such as Bruggeman's effective medium theory [29], or the random fiber model by Tomadakis and Sotirchos [30]. The deviations from these idealized models are explained by the complex structure formed by fibers, binder, and PTFE in carbon-paper materials. For instance, the sintering processes of carbon-fiber GDLs typically results in a webbed region at fiber intersection points that offers little porosity but dramatically increases tortuosity.

In contrast, very few experimental measurements on the relative effective diffusivity, $g(s)$, of GDLs have been reported in the literature [10,31,32]. This is due to the difficulty of measuring species concentration and flux in GDLs with thicknesses between 100 and 400 μm , while simultaneously controlling the amount of water. For instance, Utaka et al. [31] used custom GDLs, which were 10–20 times thicker than those actually used in fuel cells, in order to achieve a detectable concentration gradient. Hence, a

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