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Alternative analytical analysis for improved Loschmidt diffusion cell

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ABSTRACT

To measure gas phase diffusion coefficients across porous media, an apparatus called a Loschmidt diffusion cell is often utilized. In previous studies with such an apparatus, an infinite-length assumption is used to simplify the analytical solution. Experimentally, cell lengths must be quite long and measurement time is very brief to fulfill this assumption. In this study, Fick's second law is applied, and separation of variables with shifted homogeneity technique is performed for data analysis to enable design of a more compact experimental apparatus with extended measurement times and improved precision. The analytical solution is proved by both the inverse-matrix method and finite-volume discretization. Finally, using the new analytical solution obtained, the effective diffusion coefficient is determined for porous media used in fuel cell applications.

1. Introduction

A Loschmidt diffusion cell is a combination of two gas chambers with a removable barrier between them. This apparatus is used for determination of binary and effective diffusion coefficients of gases flowing through porous media. Initially, each chamber contains different gas concentrations. Due to the concentration difference, when the barrier is removed, gas is exchanged until equilibrium is reached. The concentration change at a point as a function of time can be used to determine the diffusion coefficient. Detailed information about Loschmidt diffusion cells can be found in [1].

There are a limited number of published studies about determination of binary and effective gas diffusion coefficient through porous media in fuel cells using a Loschmidt diffusion cell. In the polymer electrolyte fuel cell (PEFC), several types of porous media exist, including the gas diffusion layer (GDL), micro-porous layer (MPL) and the catalyst layer (CL) (Ref. [2, p. 50]). Diffusion transport is critical in these media, which may also be partially saturated.

Göll and Piesche [3] used a macroscopic approach and presented a new computational model for diffusion in porous media. An isothermal diffusion problem in a Loschmidt tube simulation was used for validation of their new model. Shen et al. [4] used the Loschmidt cell to measure the effective gas diffusion coefficients of a dry O_2–N_2 gas pair in dry fuel cell cathode catalyst layers at 25°C. Astrath et al. [5] investigated the effective gas diffusion coefficients of four different types of stainless steel films with different shaped holes using a Loschmidt diffusion cell. Zamel et al. [6] performed experimental determination of the effective diffusion coefficient of an O_2–N_2 gas mixture through Toray TPGH-120 GDL for fuel cells via a Loschmidt cell. A Loschmidt cell was used for the experimental measurement of effective diffusion coefficient through a GDL MPL composite by Chan et al. [7]. Effective gas diffusion through SGL Sigracet 10-series GDL with 0% and 5% PTFE loading without MPL coating and 25-series GDL with 20% PTFE and MPL coating were analyzed. Zamel et al. [8] developed a correlation for the effective gas diffusion coefficient in carbon paper. An in-house Loschmidt cell with a photothermal-deflection probe was employed to measure the effective diffusion coefficient of a GDL with 70% porosity with a CO_2–O_2 gas mixture by Rohling et al. [9].

In all these studies, the series solution, which is introduced in Refs. [10,11], is used for the diffusion coefficient calculations. In this study, Fick's second law of diffusion is applied to the diffusion problem involving a Loschmidt diffusion cell with a porous layer. The analytical solution is proved by both the inverse-matrix method and finite-volume discretization. Finally, by using the obtained analytical solution, the effective diffusion coefficient is determined for a porous sample.

The previous studies referenced Crank [10] and Carslaw and Jaeger [11] for the derivation of the analytical solution of Fick's second law of diffusion. In this study, Haberman's [12] solution to one-dimensional diffusion problems with non-homogenous

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Nomenclature

A	constants for analytical analysis
a,b,c	coefficients of series
C	oxygen concentration
D	diffusion coefficient
D_{bulk}	binary diffusion coefficient of O_2 and N_2
D_{eq}	equivalent diffusion coefficient
D_{eff}	effective diffusion coefficient
J	mass flux
l	porous sample thickness
L	channel length
M	constants for inverse-matrix analysis
Q	diffusion ratio
t	time
Δt_{expl}	critical time step for CFL condition
z	cell direction axis
Z	distance between the gate and the oxygen sensor

Greek Letters

α,β,ξ	variables, which are used for the simplification of the results
λ	eigen value
φ	interface oxygen concentration

Subscripts

1	Region 1
2	Region 2
i	position index
n	time index
k	summation index
p	porous region

Superscripts

n	time steps

boundary conditions is followed. To the authors' knowledge, there is no study found in literature that deals with nonhomogeneous case of diffusion media. This method allows a short, more compact experimental apparatus and provides greater accuracy through extended measurement times.

2. Experimental design

A simple sketch of this cell used is shown in Fig. 1. The half-length of the channel is 0.08 m and the inner diameter of the cell is 0.026 m. A ball valve is located between the two chambers and a Sigracet SGL 10AA porous sample was inserted in one chamber near the valve to investigate the effective diffusion coefficient. There is about 200 µm distance between the valve and the sample. To simplify the calculations; this gap is assumed to be on the lower side of the valve (in the first chamber); hence the distance between the valve and the porous sample is assumed to be zero.

For the experiments, first, the upper chamber of the cell is purged with a gas mixture of 90% nitrogen with 10% oxygen, whereas the lower chamber contains atmospheric air. After equilibrium is obtained and the homogeneity is observed, the initial oxygen content in both chambers is measured. Then, the valve between the chambers is opened to enable gas diffusion and the oxygen molecules diffuse from the lower chamber to the upper chamber. The dynamic O_2 concentration is measured and recorded for about three minutes with a measurement frequency of 2 Hz, by an Ocean Optics Neofox 1000 µm oxygen sensor, which is placed in the upper chamber, at a location of 13.5 mm from the valve.

The governing equation for mass transfer in the cell is "Fick's second law" and can be seen in Eq. (1).

$$\frac{\partial C}{\partial t} = D\frac{\partial^2 C}{\partial z^2} \tag{1}$$

In this equation, D becomes D_{eff} if the diffusion medium is porous. If the medium is open, then D becomes D_{bulk}. By using the measured concentration at the sensor point and the analytical solution of the Fick's second law, D_{eff}/D_{bulk} values are deduced.

Two different methods were used to obtain the analytical solution of the Fick's second law. First, the regular solution, which is called "the standard method" in the current study, was applied

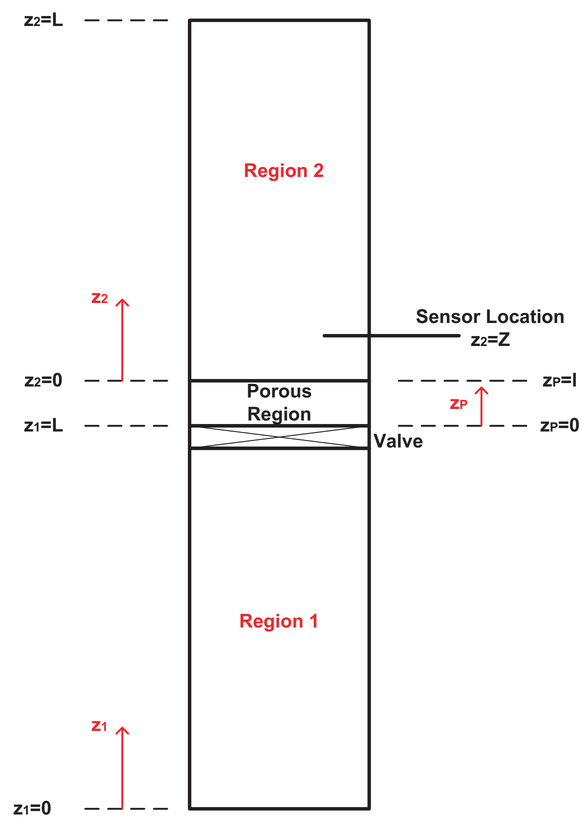

Fig. 1. A simple schematic of the Loschmidt diffusion cell.

for the experiments. Then a new analytical solution, which is called "the new method" was utilized. This new solution method is introduced in Section 3,

For the case with the standard solution; the following equations (Refs. [4–9]) were applied and the effective diffusion coefficients were determined.