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A method for measuring in-plane effective diffusivity in thin porous media

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ABSTRACT

A new experimental technique for measuring the in-plane components of the effective diffusivity tensor of thin porous materials is presented. The method is based on the transient diffusion of oxygen from air into a porous sample initially purged with nitrogen. The oxygen concentration is measured at a fixed location in the sample with time and the response is fitted to an analytical solution of Fick's law for one-dimensional, transient diffusion. As validation, it was confirmed that this method reproduced the theoretical value of oxygen diffusivity in nitrogen within 1% when no sample is present. Effective diffusion coefficients were measured for a variety of thin fibrous graphite paper materials typically used in fuel cell electrodes. The sample holder was designed to allow varying degrees of compression, thereby changing the porosity and tortuosity of the material. As expected the effective diffusivity drops with compression, not only due to a decrease in porosity but also to a large increase in tortuosity. The present method provides accurate, fast, and repeatable measurements, is applicable to electrically conductive materials where brine conductivity is difficult to interpret, uses a simple sample holder, an off-the-shelf oxygen sensor, and involves only air and nitrogen gas. The obtained values were in excellent agreement with comparable results in the literature, yet with a much more direct method.

1. Introduction

Global fossil energy resources are expected to be largely depleted within our lifetime [1]. This troubling fact, in addition to their contribution to the emission of greenhouse gases, has motivated the search for alternative fuels and energy sources. The hydrogen fuel cell is a major component of this vision, since hydrogen can be easily produced from many different methods, such as reforming natural gas or biogas [2,3], electrolyzing water using wind power or solar farms [4], or even splitting molecular water using solar powered photolytic reactions [5]. Hydrogen fuel cells, also known as Polymer Electrolyte Membrane Fuel Cells (PEMFCs), are particularly promising for mobile and automotive applications due to their high power density and quick refueling times, comparable to internal combustion engines. Most major automotive manufacturers have committed to offer fuel cell vehicles commercially between 2015 and 2020 [6].

Fig. 1 illustrates a schematic cross-section of a fuel cell assembly showing the flow field plates, the gas diffusion layer (GDL), catalyst layer (CL), and the polymer electrolyte membrane (PEM). Also shown schematically in Fig. 1 are the many transport mechanisms that occur simultaneously through the various porous components during cell operation. A detailed description of PEM fuel cell operation can be found elsewhere in review articles [7,8] and textbooks [9,10]. One of the more important transport processes is the diffusion of gaseous reactants from the flow channels through the GDL to the CL. The GDL plays many roles inside the PEMFC, including conduction of heat and electrons and provision of mechanical support to the soft membrane; however, as the name suggests, their primary purpose is to disperse gaseous reactants from the flow channels to regions of the catalyst layer under the ribs. The rate at which gas diffuses through the GDL is directly linked to the amount of electric current generated, but also impacts the efficiency of cell operation through the phenomena of concentration polarization [4]. It is consequently of great importance to properly characterize the gas diffusivity of these materials, with the aim of reducing mass transport limitations and increasing fuel cell efficiency. Engineering the fuel cell to operate at higher current density means that cells can be made smaller and more cost-effective for a given power rating, and operating at high efficiency means longer ranges between refilling.

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Nomenclature

Symbol	Description
A	area (m^2)
C	concentration (%)
D	diffusion coefficient ($cm^2\ s^{-1}$)
D_b	bulk diffusion coefficient ($cm^2\ s^{-1}$)
D'	normalized effective diffusivity (–)
l	length domain (m)
n	molar flow rate ($mol\ s^{-1}$)
t	time (s)
z	spatial coordinate (m)

Greek symbols

δ	sample thickness (m)
ε	porosity (–)
τ	tortuosity (–)

Subscripts

0	initial
1	final
eff	effective

There are two main difficulties with measuring the effective diffusivity in the GDL. Firstly, the materials are very thin, making it challenging to apply controlled boundary conditions. For instance, their high permeability combined with their minimal thickness mean that even slight pressure differences cause significant convective flows. Secanell and co-workers [11] have developed a Wicke–Kallenbach type cell for measuring through-plane (TP) diffusivity, but this was only feasible for materials with a microporous layer (MPL), whose low permeability buffered against pressure fluctuations. Even so, this type of setup requires very careful control of the pressure, composition, and flow rates on each face of the sample. The second difficulty is that GDLs are electronically conductive. Therefore, the standard porous media approaches based on measuring brine conductivity to infer formation factor [12,13] will not work directly, as the ionic and electronic transport must be accounted for. Büchi and co-workers [14] have developed a sophisticated technique using electrochemical impedance spectroscopy to de-convolute the effects of these two transport mechanisms. Not only is this method somewhat complex and difficult to reproduce for non-electrochemists, it does not actually measure effective diffusivity directly.

A variety of other experimental approaches have been taken in attempts to study the diffusivity in GDL. Astrath et al. [15] developed a Loschmidt method [16] to study diffusion through porous separators and membranes, and this group subsequently studied the TP effective diffusivity of GDL materials [17]. This approach involves allowing two separate gas chambers with differing initial concentrations to counter-diffuse into each other. When the chambers are separated by the GDL, gases must diffuse through it, thereby adding a diffusive resistance to the process. Quick et al. [18] and LaManna et al. [19] developed an experimental technique to measure the effective diffusivity of water vapor, by creating a humidity gradient in the porous sample via a dry and a humidified flow channel. A similar approach was used by Baker and co-workers [20] but without flow. Utaka et al. [21] built an electrochemical oxygen sensor to measure the effective diffusion coefficient of microporous media under dry and wet conditions. This technique was interesting, since the electrochemical oxygen sensor consumed oxygen to establish a stable concentration gradient through the sample. Moreover, the current produced by the sensor indicated the flux, and the voltage of the sensor provided the concentration. Their setup required manufacturing a custom

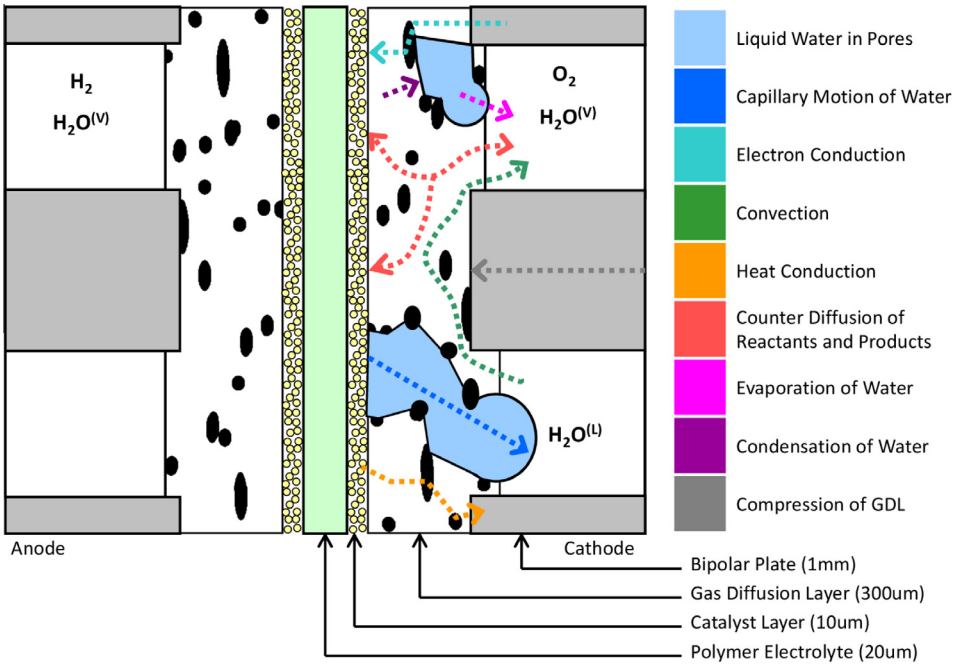

Fig. 1. Schematic of a PEMFC assembly illustrating mass transport and phase change mechanisms inside a fuel cell.