



Theoretical examination of effective oxygen diffusion coefficient and electrical conductivity of polymer electrolyte fuel cell porous components



Gen Inoue^{a,*}, Kouji Yokoyama^b, Junpei Ooyama^b, Takeshi Terao^b, Tomomi Tokunaga^b, Norio Kubo^b, Motoaki Kawase^a

^a Department of Chemical Engineering, Faculty of Engineering, Kyoto University, Kyoto-daigaku-katsura, Nishikyo-ku, Kyoto 615-8530, Japan

^b Fuel Cell Cutting-Edge Research Center Technology Research Association (FC-Cubic TRA), 2-3-26, Aomi, Kouto-ku, Tokyo 135-0064, Japan

HIGHLIGHTS

- Actual porous electrode structures of PEFC were made with FIB-SEM and X-ray CT.
- Both gas diffusion results of calculations and experiments were almost same.
- Equations of relative diffusion coefficient of each porous media were obtained.
- Catalyst layer had low gas diffusion property depending on ionomer as expected.

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ABSTRACT

The reduction of oxygen transfer resistance through porous components consisting of a gas diffusion layer (GDL), microporous layer (MPL), and catalyst layer (CL) is very important to reduce the cost and improve the performance of a PEFC system. This study involves a systematic examination of the relationship between the oxygen transfer resistance of the actual porous components and their three-dimensional structure by direct measurement with FIB-SEM and X-ray CT. Numerical simulations were carried out to model the properties of oxygen transport. Moreover, based on the model structure and theoretical equations, an approach to the design of new structures is proposed. In the case of the GDL, the binder was found to obstruct gas diffusion with a negative effect on performance. The relative diffusion coefficient of the MPL is almost equal to that of the model structure of particle packing. However, that of CL is an order of magnitude less than those of the other two components. Furthermore, an equation expressing the relative diffusion coefficient of each component can be obtained with the function of porosity. The electrical conductivity of MPL, which is lower than that of the carbon black packing, is considered to depend on the contact resistance.

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1. Introduction

Achieving further cost reductions in Polymer Electrolyte Fuel Cells (PEFCs) not only requires the development of new materials such as non-platinum catalysts and low-platinum technologies, but also overcoming the extremely important challenge of increasing current densities [1]. The realization of high current densities is generally expected to make it possible to reduce the electrode area

on the PEFCs stack, thereby reducing the amount of material required and lowering the cost. In present PEFCs, the reactions and transport resistance associated with the oxygen reduction reaction (ORR) at the cathode are the rate-limiting factors, and many studies have focused on the cathode [2–6]. The crucial function of the cathode is the requirement to supply oxygen, protons, and electrons rapidly and smoothly to the Pt electrode. Fig. 1 shows a schematic diagram of the cathode oxygen reduction reaction, oxygen transport at the cathode, and SEM images of various porous media. In particular, an insufficient supply of oxygen to the electrode surface can obstruct the achievement of high current densities; thus, improvements to the oxygen transport rate are

* Corresponding author.

E-mail address: ginoue@cheme.kyoto-u.ac.jp (G. Inoue).

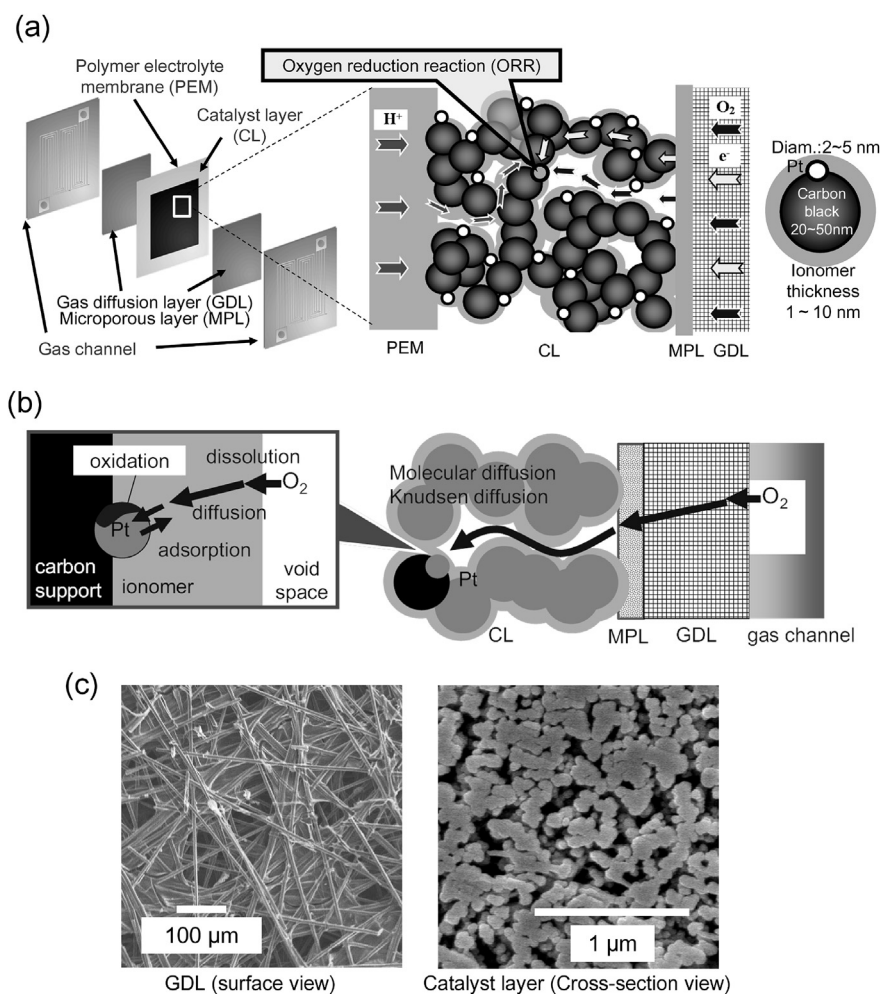


Fig. 1. Schematic diagram of (a) cathode oxygen reduction reaction, (b) oxygen transport at the cathode, (c) SEM images of various porous media.

essential. Guided by the illustration in Fig. 1(b), improvements are needed in several areas: the oxygen diffusion rate from the gas flow channel through “macro-pores” or “meso-pores” in the gas diffusion layer (GDL), microporous layer (MPL), and catalyst layer (CL); the rate of micro-scale diffusion to ionomers, the production of liquid water and the presence of “micro-pores” in the carbon support; and the adsorption rate onto the Pt electrode surface. In relation to mass transport in porous media, pore diameters in the field of catalyst chemistry are defined as follows by the International Union of Pure and Applied Chemistry (IUPAC): less than 2 nm, “micro-pore,” from 2 to 50 nm, “meso-pore,” and larger than 50 nm, “macro-pore.” The former type of micro-scale mass transport phenomena has been the subject of a considerable amount of research and numerous development studies [7–10], such as those involving ex-situ measurements and calculations that are based on molecular dynamics and models of the ionomer coating structure to understand the resistance mechanism and improve transport resistance. On the other hand, the latter type of macro-level mass transport is also an extremely important component influencing the overvoltage of the cell; hence, reducing the transport resistance of this process is essential. Fig. 1(c) shows the porous structure of each component. Neglecting the resistance due to Knudsen diffusion, the effective diffusion coefficient D^{eff} for a porous medium may be expressed as a function of the parameters of the porous structure—its bulk molecular diffusion coefficient D^{bulk} , the porosity ϵ , and tortuosity according to equation (1). The tortuosity

measures the curved length of transport paths within the pores, where large tortuosity values signify twisty, winding curves. The smallest possible value of the tortuosity is 1, which corresponds to a straight-line path.

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

This equation clearly indicates that increasing the mass transport rate, which improves the effective diffusion coefficient, requires a structure with high porosity and low tortuosity. Because we require electronic conductivity within the acidic environment of a proton-conducting PEFC, carbon materials are the primary choice of material for the structural frameworks of the GDL, MPL, and CL. The most common type of GDL is a fibrous porous media (paper structure) with a thickness of roughly 200 μm and a porosity of approximately 0.8, produced from carbon fibers with a diameter of 10 μm. The mean pore diameter is of the order of 20 μm. The structure also incorporates a binder to improve the structural integrity and the contact between fibers. The MPL is an accumulated layer of carbon black (CB) possessing a primary aggregate of carbon particles with approximate diameters in the range 20–50 nm. PTFE is typically used as a binder. The porosity is roughly 0.6–0.7, the mean pore diameter is roughly 0.1 μm, and the thickness is typically in the range 10–50 μm. The CL is an accumulated layer of Pt catalysts supported on carbon black; its structure also includes ionomers (electrolytic polymers) that constitute

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