Direct simulation Monte Carlo analysis of gas transport in microporous structure based on X-ray computed tomography

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Introduction

The improvement of gas transport characteristics in porous materials is required to increase the current density of polymer electrolyte fuel cells (PEFCs). While the structure and gas transport phenomena of gas diffusion layers, which have a porous structure with a characteristic length scale of ~10 μm, were extensively investigated, those of microporous layers (MPLs) have not been well understood because of the difficulty of the measurements in sub-μm porous structures.

This paper describes the analysis of gas transport in MPLs based on the three-dimensional structure obtained from X-ray nano computed tomography (CT). We employed the direct simulation Monte Carlo (DSMC) method [1] in order to accurately analyze the gas transport phenomena where the characteristic length scale of the structure is comparable to the mean free path of gas molecules. The numerical simulation well reproduces the experimentally observed pressure dependence of diffusion resistance originating from the coexistence of Knudsen and molecular diffusion mechanisms. The effect of porous media morphology on gas transport was examined by an analysis of molecular trajectories.

Construction of 3D structure data

A MPL sample (acetylene black : PTFE = 0.8 : 0.2) was trimmed by focused ion beam milling and the three dimensional structure was obtained from X-ray scans (Xradia UltraXRM-L200) [2]. The volumetric structure data was obtained for the 5.76 μm cubic region with the voxel size of 32 nm. The polygonal surface representation of the porous structure was constructed from these images using the marching tetrahedrons algorithm [3] (Fig. 1). The threshold value for the isosurface extraction was determined so that the porosity of the generated structure agrees with the value measured by mercury porosimetry (64%).

Gas transport analysis using DSMC method

Figure 2 shows the simulation system, where the top and bottom side of the porous medium have contact with gas reservoirs of pure nitrogen and oxygen. The pressure of the reservoirs is set at between 1 – 300 kPa. The DSMC calculation employed the cells of 180 nm on each side and the variable hard sphere (VHS) model [1] for gas collisions. The cosine scattering with perfect energy accommodation was assumed for gas-surface interactions. Gas molecules were specularly reflected on the side boundaries of the system. We determined the net flux of gas molecules across the porous structure from the difference between the inflow flux at the top and bottom boundaries given as the boundary condition and the outflow flux calculated in the simulation. After the system reaches the steady state, we sampled the net fluxes at the top and bottom boundaries and determined the diffusion resistance $R = \Delta n / J$, where Δn is the number density difference of gas molecules and J is the net flux of gas molecules across the porous structure.

Results and discussions

The diffusion resistance of oxygen across the porous structure is shown in Fig. 3. The simulation results are converted to values for the porous layer thickness of 45.8 μm so that we can directly compare these values with the experimental results. Our DSMC simulation well reproduces the diffusion resistance measured in the experiment. The y-intercept value corresponds to the Knudsen diffusion resistance, where gas molecules collides only with solid surface. As the pressure increases, the diffusion resistance increases because of the more frequent collisions between gas molecules.

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References

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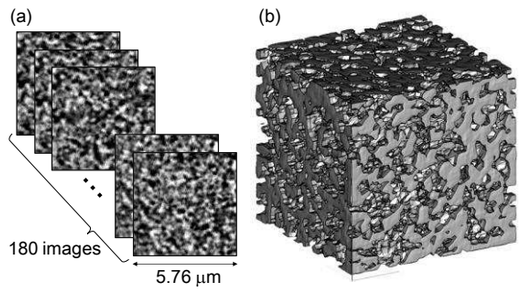

Fig. 1 Cross-sectional images obtained by X-ray nano CT (a) and the polygonal surface representation (b) of the micro porous layer.

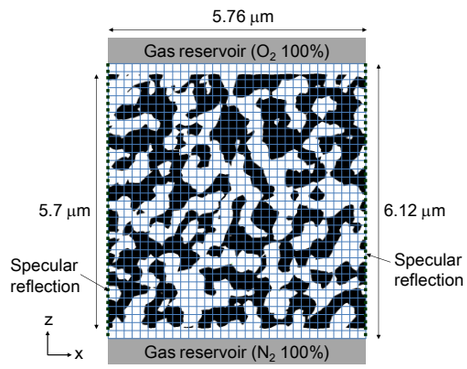

Fig. 2 Simulation system for the evaluation of gas diffusion resistance (cross-sectional view).

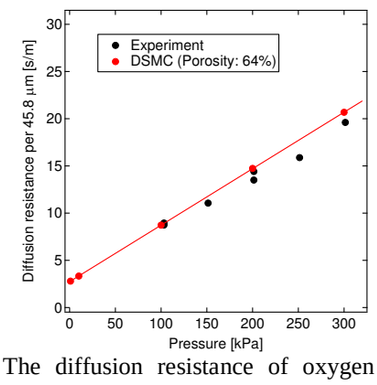

Fig. 3 The diffusion resistance of oxygen across the porous structure.