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Effect on the binder morphology on stochastic reconstruction of gas diffusion layer in PEMFC#

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ABSTRACT

The gas diffusion layer is one of the most important components of the proton exchange membrane fuel cell (PEMFC). Microstructure reconstruction is an efficient research strategy. The influence of the morphological presence of resin binder when applying a stochastic method to reconstruct the three-dimensional gas diffusion layer was studied. Finite element method (FEM) simulation was conducted to explore the mechanical response under compression load for both reconstruction models with the binder morphologically present or absent. The differences in pore structure of the two reconstructed models before and after compression were meticulously analyzed. The morphological appending of the binder made the overall equivalent modulus larger, but the deformation and stress concentration became severe. When subjected to a compression ratio of 20%, the porosity changed to 74.4% and 74.7% for structures with and without the presence of binder morphology, respectively. Binder morphological addition altered the pore space greatly, making it a more uniform pore size distribution and a smoother porosity variation. In the through-plane (TP) direction, the morphological presence of the binder decreased the tortuosity, while in the in-plane (IP) direction, the tortuosity increased.

Keywords: PEMFC, gas diffusion layer, binder morphology, mechanical behavior, pore structure

NONMENCLATURE

Abbreviations	
PEMFC	Proton exchange membrane fuel cell
GDL	Gas diffusion layer
FEM	Finite element method
MPL	Microporous layer
BP	Bipolar plate
MEA	Membrane electrode assembly
CL	Catalyst layer
IP	In-plane
TP	Through-plane

1. INTRODUCTION

Hydrogen energy is one of the most promising energy sources in the future for its zero-emission, high efficiency, and wide application range[1, 2]. PEMFC is an important utilization terminal for hydrogen energy, whose key components include proton exchange membrane (PEM), catalyst layer (CL), gas diffusion layer (GDL), and bipolar plate (BP)[3]. GDL provides mechanical support for the membrane electrode assembly (MEA), conducts heat and charge, diffuses reaction gas, and expels product water[4].

GDL consists of a macroporous substrate and a microporous layer (MPL), which will be compressed during the fuel cell assembly process[5, 6]. Carbon paper is a widely used commercial substrate and has attracted much research attention[7, 8]. Carbon paper comprises carbon fiber reinforcement and resin matrix[9]. The arrangement of fibers and the distribution of binders are too random to be analyzed quantitatively[10], making it challenging to predict the internal structure changes when subjected to compression load. GDL microscale reconstruction constitutes an effective method to address this problem[11, 12]. Specifically, the stochastic model has the advantages of being flexible and cost-effective[13]. Mang researchers have conducted FEM simulations to investigate the compression process based on the stochastic reconstruction model[14, 15]. The diverse constructions exert significant influence on the mechanical properties and mass transfer capacity[16, 17].

For simplification, many researchers ignore the binder's morphological presence and treat it as bonded contact constraints with carbon fiber[18, 19]. Few research has been reported on the impact of the morphological presence or absence of the binder on the mechanical properties and pore space structure when

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applying reconstruction methods. On the other hand, the binder morphology can be coupled to the reconstructed fiber skeleton in various ways to rebuild the carbon paper microstructure more accurately[19-21].

In this paper, the stochastic reconstruction method was used to obtain the carbon fiber porous structure. The morphological presence of binders was considered based on the opening operation. The different mechanical properties for both reconstruction models appended binders morphologically or not were studied using FEM for the first time, revealing the influence of binder morphology on compressibility. The pore space characteristics for the two cases were compared overall. This study provides a reference for whether to consider the presence of binder morphology when stochastically reconstructing carbon paper GDL.

2. MODELING AND SIMULATION

Fig. 1 exhibits the morphology of paper-type GDL without hydrophobic treatment. The carbon fibers are randomly oriented in a plane, leading to different IP and TP properties. The resin carbon adheres to the surface of carbon fibers and inclines to aggregate at the intersections of carbon fibers, presenting a disk shape.

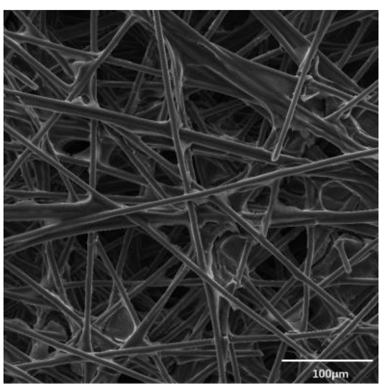

Fig. 1 SEM image of paper-type GDL.

In this paper, a stochastic generation method was applied to reconstruct the microscopy porous structure of the GDL. Before considering the presence of the binder, the carbon fiber skeleton structure was acquired based on the following assumptions for simplification: (i) carbon fibers were straight cylinders with a fixed diameter; (ii) fibers were set as finite length but significantly longer than the domain size; (iii) overlapping between carbon fibers was allowed. The fibers were generated iteratively until the target porosity was reached. In one case, the resin binder was morphologically ignored and treated as uniformly distributed along the fibers. On the other hand, the morphological presence of the binder was considered via the opening calculations, which were implemented based on mathematical morphology and defined as corrosion followed by expansion. In stochastic reconstruction, 0 represents the pore space, while 1 represents the solid material, and the opening operation was conducted on the binary three-dimensional matrix[22]. Fig. 2 shows the reconstructed model of paper-based GDL. Obviously, for the identical porosity, in the reconstructed model considering the binder morphologically, the quantity of carbon fibers is much lower than that in the model binder morphologically absent. For both cases, the porosity is 79%, the carbon fiber diameter is 8 μm, and a domain size of 200μm × 200μm × 190μm was selected to be representative elementary volume (REV). The geometries were then discretized for finite element method (FEM).

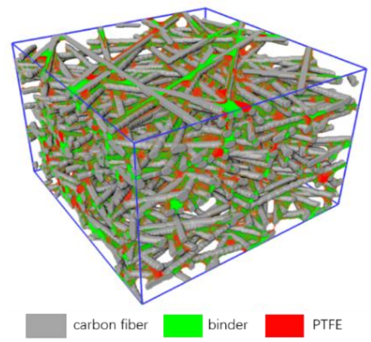

Fig. 2 Reconstruction of the macroporous substrate.

The elastic mechanics model was used to simulate the compression dynamics under the compression ratio of 20%, which is governed by the equilibrium, geometric, and constitutive equations, as listed in Eqs. (1)-(3).

Equilibrium equations:

$$\sigma_{ij.j} + \rho f_i = \rho \ddot{u} \tag{1}$$

Geometric equations:

$$\varepsilon_{ij} = \frac{1}{2}(u_{i.j} + u_{j.i}) \tag{2}$$

Constitutive equations:

$$\begin{cases} \sigma_{ij} = \lambda \varepsilon_{kk} \delta_{ij} + 2\mu \varepsilon_{ij} \\ \sigma_{kk} = (3\lambda + 2\mu)\varepsilon_{kk} = 3E\varepsilon_{kk} \end{cases} \tag{3}$$

Where σ is stress, ε is strain, ρ is density, f is body force, u is displacement, $\ddot{u}$ is acceleration, λ and μ are the Lame constants, δ_{ij} is the Kronecher symbol, and E is Young's modulus.

For simplification, the binder and carbon fibers were treated as isotropic solid materials with Young's modulus of 300MPa, Poisson's ratio of 0.256, and density of 1800kg/m³. The MPL and BP were regarded as rigid bodies.

The tortuosity is the square of the actual diffusion distance divided by the Euclidean distance of the opposite surfaces: $\tau=(L_{act}/L)^2$. Tortuosity can also be defined as follows:

$$\tau = \varphi \frac{D}{D_{eff}} \quad (4)$$

where D is the intrinsic diffusion coefficient, and ϕ is the porosity. Effective diffusivity can be obtained from the pore scale model by solving Fick's law as shown:

$$D_{eff} = \frac{N_A L}{A \Delta C} \quad (5)$$

3. RESULTS AND DISCUSSION

Fig. 3 illustrates the influence of whether the binder presents morphologically or not on the mechanical properties of GDL. Furthermore, the validation research between FEM and the experiment was conducted. Both the two cases and the experiment show the nonlinearity of stress-strain curves for carbon paper subjected to compression, the reason for which is the increasing contact points and contact area. The simulated mechanical response of the case ignoring the binder morphologically is aligned with the compression experiment. However, the equivalent elastic modulus of the case considering the binder morphologically is much higher than that of the case in which the binder is morphologically absent, namely the former porous structure is harder to deform. Although the number of carbon fibers in the model considering binder morphologically is relatively low, the binder at the intersections reinforces the mechanical strength of otherwise loose fiber structure. The presence of the binder transforms the internal solid parts of the carbon paper from point and line contacts to surface contacts under compression, thereby increasing the area resisting deformation. Besides, the deterioration of the binder, such as fracture and detachment from carbon fibers, was not studied during the simulation, making the stiffness exceed the actual value.

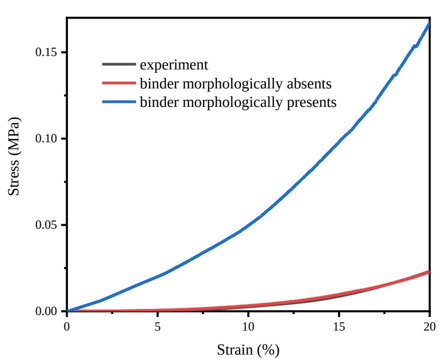

Fig. 3 Stress-strain curves for simulation and experiment.

Fig. 4 illustrates the deformation and stress distribution for the two reconstruction cases. For the model containing binder morphologically, the maximum value for both displacement and stress is apparently higher, inferring that the deformation and stress concentration are more severe. Due to the longer distance between adjacent fibers and the strong bond of the binder, carbon fibers suffer severe bending.

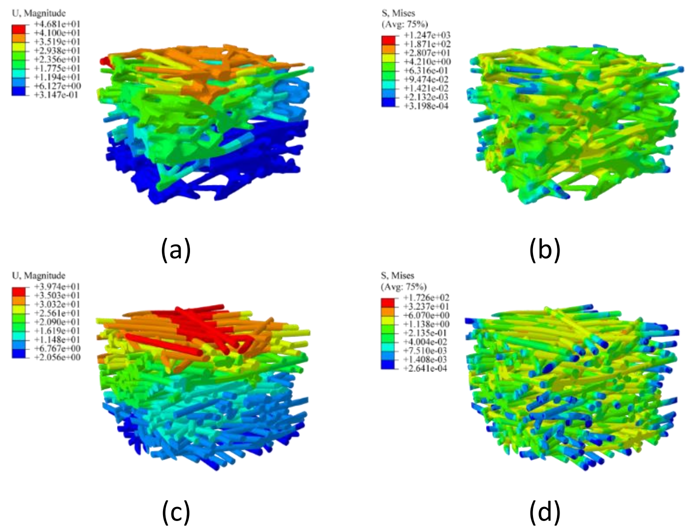

Fig. 4 Displacement and stress distribution:(a) displacement in reconstruction with binder morphology, (b) stress in reconstruction with binder morphology, (c) displacement in reconstruction without binder morphology, (d) stress in reconstruction without binder morphology.

Fig. 5 shows the normalized porosity distribution along the thickness direction and pore size distribution for the reconstruction models before and after compression. The porosity of the model with binder morphologically or not decreases to 74.7% and 74.4% at the compression ratio of 20%, respectively. The porosity variation along the through-plane direction for the porous medium considering the binder morphologically is smoother with less fluctuation. When the reconstructed model is without a binder morphological present, the small pore dominates. In contrast, when the binder is appended morphologically, the pore size shifts towards larger pores and becomes more uniform. This indicates that binder presence has an important influence on the pore space when modeling the GDL microstructure. Besides, regardless of whether the binder is appended morphologically, under compressive load, the pore size distribution shifts towards smaller pores, with a decrease in the number of large pores and an increase in the number of small pores.

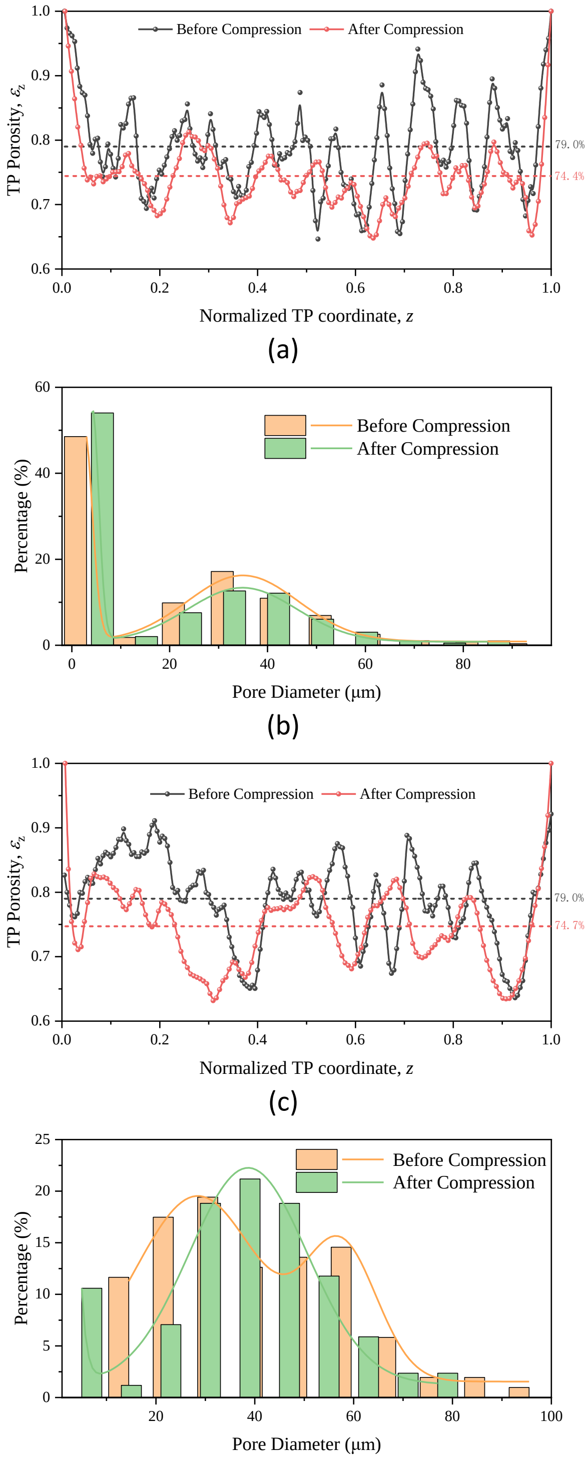

Fig. 5 Porosity variation along thickness and pore size distribution for (a) (b) reconstruction without binder morphology, (c) (d)reconstruction with binder morphology.

As for the mass transfer capacity variation shown in Fig.6, no matter whether the binder is appended morphologically, in the TP direction, the tortuosity increases, and effective diffusivity decreases under 20% compression. Due to more uniform pore size distribution, the tortuosity of the model containing binder morphologically is lower than that of the other model, thus leading to a better mass transfer in the TP direction. However, the mass transfer in the IP direction faces greater obstruction for the model with binder morphologically. The reason is that the opening operation introduced more binder that connects interlayer carbon fibers into the construction of the carbon paper. In the case without binder morphology, the fibers are oriented parallel to the IP direction, resulting in less tortuosity. After compression, the sharp decrease of the tortuosity in the IP direction leads to a prominent growth of effective diffusivity. When the tortuosity changes are minimal, the effective diffusivity would be more affected by the overall porosity.

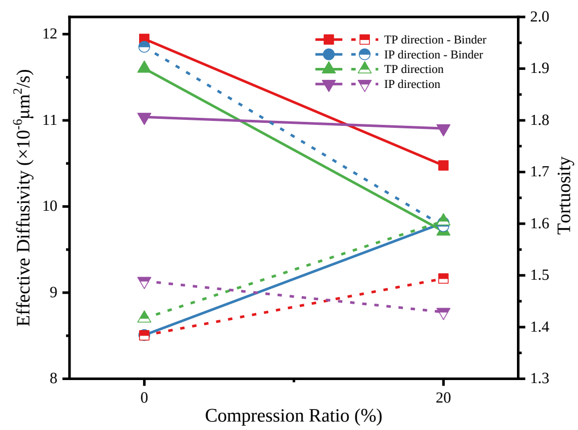

Fig.6 Tortuosity and effective diffusivity variation under compression

4. CONCLUSIONS

The influence of the morphological presence of binder during the reconstruction of GDL was studied in this research. FEM illustrates that both the reconstructed porous medium with the binder morphologically appended or not exhibit mechanical nonlinearity response when subjected to compression load. The model considering the binder morphologically present yields a larger equivalent elastic modulus compared to the other model morphologically ignoring the binder and experiment result, and its deformation and stress concentration are severer. The reconstruction with binder morphologically appended has a more uniform pore size distribution and smooth porosity variation along the thickness, while a concentrated distribution of small pores appears in the reconstruction with binder morphologically absent. The addition of binder decreases the tortuosity along the TP direction but increases it along the IP direction. The morphological presence of the binder alters the trend of mass transfer capability in carbon paper after compression.

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