Numerical Determination of Transport Properties of Catalyst Layer, Microporous Layer and Gas Diffusion Layer

Jürgen Becker, Math2Market GmbH, Kaiserslautern

10th Symposium for Fuel Cell and Battery Modelling and Experimental Validation

Bad Boll  March 19-20, 2013
Math2Market GmbH

- M2M is a spin-off from the Fraunhofer Institute for Industrial Mathematics ITWM
- M2M was founded in Sep. 2011 by 3 developers of the GeoDict software.
- M2M has acquired all rights for marketing and developing GeoDict from Fraunhofer.
- M2M is located in the „Innovationszentrum Westpfalz“ at Kaiserslautern.
- Close cooperation with Fraunhofer ITWM.
GeoDict: The Virtual Material Laboratory ...

- Import
- Analyze Geometry
- Create
- Determine Properties
... Applied to Porous Transport Layers of PEFC

1. Determine Transport Properties from 3D Images
   a. GDL Analysis Based on Tomography Image
   b. Catalyst Layer Model Based on FIBSEM Images

2. Create 3D Structure Models Virtually
   a. GDL Model: Fibers, Binder, Compression
   b. MPL Design Study
1. Determine Transport Properties from 3D Images

a. GDL Analysis Based on Tomography Image
Tomography Image

Input: tomography image
- Carbon fibres of diameter ~ 7 µm
- Hydrophobic PTFE coating
- Porosity 78%
- Layer thickness ~ 200 µm
- Picture shows area of size 717x717 µm
- Resolution: 0.7 µm/voxel

Aim:
- Find capillary pressure curve, relative permeability, relative diffusivity
Permeability

Macroscopic description (homogenized porous media model)

Darcy’s law: \[ u = \frac{1}{\mu} \kappa \nabla p \]

- \( u \): average flow velocity
- \( \kappa \): permeability tensor \textit{unknown}
- \( \mu \): viscosity
- \( p \): pressure

Microscopic description (pore structure model)

Stokes equation: \[-\mu \Delta u + \nabla p = 0\]

Boundary conditions: no-slip on fibre surface, pressure drop \( \kappa \) can be determined from the solution!
Relative Permeability

Two-step approach:

1. Use pore morphology method (Hilpert, 2001) to determine distribution of air and water phase.
   - Idea: a pore is filled with the non-wetting fluid (=water), if \( p_c \geq \frac{2\sigma}{r} \cos \beta \)
   - Drainage and imbibition (connectivity to reservoir)
   - Residual water or air (connectivity of pores)

2. Solve Stokes equation on the remaining pore space to determine wetting phase (=air) permeability.

Relative Permeability

non-wetting fluid

wetting fluid

\( r \)

\( \beta \)
Capillary Pressure Curve

Parameters:
- Contact angle: 140°

Results:
- Bubble point (drainage): 8.8 kPa
- Saturation at bubble point: 20.8%
Relative Permeability
Water Distribution at Bubble Point

\[ p = 10.6 \text{ kPa} \quad (r=10.5 \text{ mm}) \]
1. Determine Transport Properties from 3D Images
   
b. Catalyst Layer Model Based on FIBSEM Images
   
   i. Reconstruction
   ii. Simulations on 3D data
i. Reconstruction from SEM Images

SEM by IMTEK, Uni Freiburg
Reconstruction Algorithm

1. Place holes randomly
2. Place particles randomly (not inside holes)
3. Fill small pores between particles
Optical Comparison
Variation of Porosity

73%  
65%  
57%
Conductivity and Diffusivity

<table>
<thead>
<tr>
<th>Porosity</th>
<th>Relative Diffusivity</th>
<th>Relative Conductivity</th>
</tr>
</thead>
<tbody>
<tr>
<td>73 %</td>
<td>51.8 %</td>
<td>7.1 %</td>
</tr>
<tr>
<td>65 %</td>
<td>38.1 %</td>
<td>13.4 %</td>
</tr>
<tr>
<td>57 %</td>
<td>25.6 %</td>
<td>21.1 %</td>
</tr>
</tbody>
</table>
ii. Simulations on 3D FIBSEM Data

- Pore Structure obtained from FIBSEM Data (IMTEK, Uni Freiburg)
- Cannot distinguish between Ionomer and Carbon
Results

Pore Size Distribution

Concentration field from diffusion simulations

Determine:
- Pore size distribution, diffusivity

2. Create 3D Structure Models Virtually

a. GDL Model: Fibers, Binder and Compression
Gas Diffusion Layer Model

Created with a stochastic process

Input:
• Porosity
• Fiber diameter and type
• Anisotropy
• (Fiber crimp)
• (Weight% binder)
Compression

Aim: how does the structure change due to clamping pressure?

Current development together with Fraunhofer

- transverse isotropic elastic modulus for fibers
- isotropic elastic modulus for binder
- 30% compression

10 min on Laptop
13.5 mio grid points
2. Create 3D Structure Models Virtually

b. MPL Design Study
MPL Design Study

- Step 1: Create model of MPL structure, determine effective parameters for MPL alone
- Step 2: Create model for GDL+MPL, determine diffusivity and conductivity of combined layer
- Step 3: What changes when we change MPL design parameters?

Comparison of Different MPL

Create MPLs with

- same porosity & carbon particle sizes
- different pore size distributions
Comparison of Different MPL

<table>
<thead>
<tr>
<th>Case</th>
<th>Conductivity</th>
<th>Diffusivity</th>
</tr>
</thead>
<tbody>
<tr>
<td>I</td>
<td>0.094</td>
<td>1.92</td>
</tr>
<tr>
<td>II</td>
<td>0.120</td>
<td>1.78</td>
</tr>
<tr>
<td>III</td>
<td>0.104</td>
<td>1.65</td>
</tr>
<tr>
<td>IV</td>
<td>0.092</td>
<td>1.59</td>
</tr>
<tr>
<td>V</td>
<td>0.095</td>
<td>1.67</td>
</tr>
</tbody>
</table>

Diffusivity:

Conductivity:
Variation of MPL Porosity
Summary: Material Properties

GDL:
- (saturation dependent) diffusivity, (saturation dependent) permeability, electric conductivity, heat conductivity
- pore size distribution, capillary pressure

MPL:
- (Knudsen) diffusivity, electric conductivity, heat conductivity
- pore size distribution

CL:
- pore size distribution, surface or contact areas, contact lines
- protonic conductivity, electronic conductivity, (Knudsen) diffusivity

Caveat: Results cannot be better than the 3D structure model permits.
Thank You!

GEO DICT
The Virtual Material Laboratory
www.geodict.com