# Simulating the Cathode Catalyst Layer of a PEM Fuel Cell using Lattice Boltzmann Modeling

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## Motivation

Polymer Electrolyte Membrane Fuel Cell (PEMFC)

- promising replacement for combustion engine and reduce $CO_2$ emissions
- needs platinum as catalysator (rare and expensive)

→platinum loading has to be reduced

→However: Performance limitations occur at low platinum loading. Aren't well understood yet. Further investigations needed.

## Approach

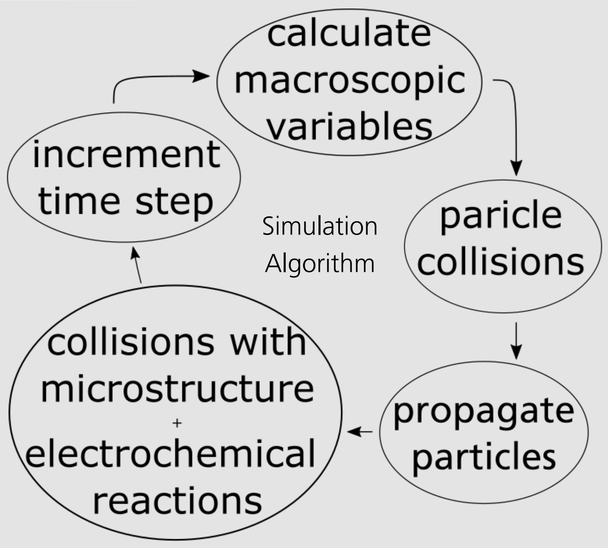
- Analyze limiting processes in cathode catalyst layer (CCL) of PEMFC on sub-µm scale
- Develop a Lattice Boltzmann Model based on [1]
- Simulate transport processes and electrochemical reactions in real CCL microstructures
- Implementation is done using the Palabos [2] framework
- Eventually, investigate influence on limiting processes in newly designed microstructures

## Simulation Model

Important processes in the CCL:

- diffusion of gases $N_2$, $O_2$, $H_2O$ in the pore space
- electrochemical reaction $4\,H^+ + 4\,e^- + O_2 \rightarrow 2H_2O$
- diffusion in the ionomer (polymer film)
- proton transport
- diffusion of liquid water in the pore space

(not yet included)

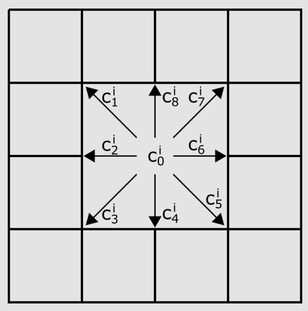



## Basic Lattice Boltzmann Model for multicomponent diffusion

Diffusion of the three gases $N_2$, $O_2$, $H_2O$ inside pore space of CCL with Lattice Boltzmann Model described in [1]:

- The Lattice Boltzmann Equation (main equation)

$$f_\sigma^i(\boldsymbol{x} + \boldsymbol{c}_\sigma^i \Delta t, t + \Delta t) - f_\sigma^i(\boldsymbol{x}, t) = \Omega_\sigma^i(\boldsymbol{x}, t)$$



discrete velocities

- Collision Operator $\Omega_\sigma^i = \Omega_\sigma^{i1} + \Omega_\sigma^{i2} + \cdots$ contains self-collisions $\Omega_\sigma^{ii}$, and cross-collisions $\Omega_\sigma^{ij}$ $(i \neq j)$:

$$\Omega_\sigma^{ii} = -\frac{1}{\tau_i}\left(f_\sigma^i - f_\sigma^{i(0)}\right), \quad \Omega_\sigma^{ij} = -\frac{1}{\tau_{ij}}\left(\frac{\rho_j}{\rho}\right)\frac{f_\sigma^{i(\mathrm{eq.})}}{c_{\mathrm{s},i}^2}\left(\boldsymbol{c}_\sigma^i - \boldsymbol{u}\right)\cdot\left(\boldsymbol{u}_i - \boldsymbol{u}_j\right)$$

- Relaxation towards equilibrium (relaxation times $\tau_i$ and $\tau_{ij}$): $\tau_i = (6\nu_i + 1)/2$

$$\left(\frac{\rho_i + \rho_j}{D_{ij}} + \sum_{s=1, s\neq i,j}^{N}\frac{n_s M_j}{D_{is}} + \frac{2nM_iM_j}{M_1 c_{\mathrm{s},1}^2 \Delta t}\right)\frac{1}{\tau_{ij}} + \sum_{s=1, s\neq i,j}^{N}\left(\frac{\rho_s}{D_{ij}} - \frac{n_s M_j}{D_{is}}\right)\frac{1}{\tau_{js}} = \frac{2\rho}{D_{ij}}$$

($\nu_i$ … kinematic viscosity, $D_{ij}$ … binary diffusion coefficients, N … number of components)
(for N<=3: analytic solutions can be found; for N>3: numeric estimations needed)

- To handle different molecular weights → rescale discrete velocities and use interpolation

## Simple simulation of electrochemical reactions

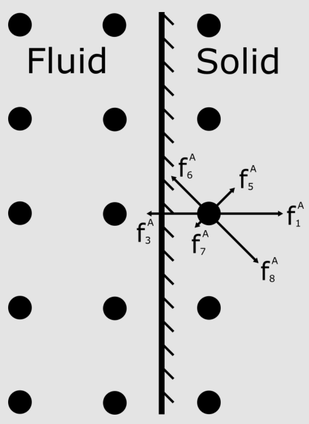
Include reaction taking place in the CCL: $4\,H^+ + 4\,e^- + O_2 \rightarrow 2H_2O$

- modified bounce-back boundary condition described in [3]
- *k* … fraction of oxygen molecules which are converted to water in each time step when in contact with platinum nano-particles
- Calculation of distribution functions (example for a right wall)

$$f_3^{\mathrm{O_2}} = (1-k) f_1^{\mathrm{O_2}}, \quad f_3^{\mathrm{H_2O}} = f_1^{\mathrm{H_2O}} + 2\frac{M_{\mathrm{H_2O}}}{M_{\mathrm{O_2}}} k f_1^{\mathrm{O_2}}$$

$$f_6^{\mathrm{O_2}} = (1-k) f_8^{\mathrm{O_2}}, \quad f_6^{\mathrm{H_2O}} = f_8^{\mathrm{H_2O}} + 2\frac{M_{\mathrm{H_2O}}}{M_{\mathrm{O_2}}} k f_8^{\mathrm{O_2}}$$

$$f_7^{\mathrm{O_2}} = (1-k) f_5^{\mathrm{O_2}}, \quad f_7^{\mathrm{H_2O}} = f_5^{\mathrm{H_2O}} + 2\frac{M_{\mathrm{H_2O}}}{M_{\mathrm{O_2}}} k f_5^{\mathrm{O_2}}$$

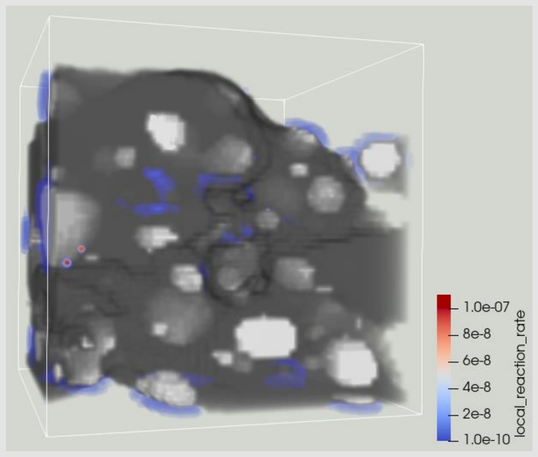


distribution functions at a right wall

## Some first results

First simulations

- of diffusion in CCL pore space with electrochemical reactions
- possible in 2D and 3D
- with real microstructure provided by the Commissariat à l'énergie atomique et aux énergies alternatives (CEA) using 3D reconstruction of binarized ADF-electron tomographic images
- concentration gradients form in nano-pores due to the oxygen reduction reaction (ORR)



local reaction rate of the ORR (red-blue scale plotted in lattice units) within microstructure (black = carbon, white = platinum)

## Outlook

Next steps:

- include diffusion in ionomer
- include Knudsen diffusion

These two effects seam to be very important.

- eventually, analyze performance limitations within different CCL mcrostructures

## Bibliography

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3. Molaeimanesh, G. R., Akbari, M. H., *Agglomerate modeling of cathode catalyst layer of a PEM fuel cell by the lattice Boltzmann method*, International Journal of Hydrogen Energy Volume **40** (2015), Issue 15, 5169 - 5185

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DLR Deutsches Zentrum für Luft- und Raumfahrt