# Effects of Agglomerate Arrangement on Cathode Catalyst Layer Performance in a PEM fuel cell

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**ABSTRACT:** Proton Exchange Membrane Fuel Cells (PEMFCs) as efficient and advantageous power sources are currently near the stage of full commercialization in the vehicle industry. However, simulations considering fuel cell cathode side are confronted with some challenges due to the complicated physics and microstructure. In the present study, reactive air flow in a cathode electrode of a PEMFC is simulated through a multi-scale lattice Boltzmann approach. In this regard, a two-dimensional single-phase lattice Boltzmann agglomerate model is developed and validated; then it is applied to model the cathode electrode consisting of a catalyst layer (CL), gas diffusion layer (GDL) and gas flow channel. To investigate the effects of agglomerate arrangement on the CL performance, species distributions in the pore region, electrical potential distribution in the electrolyte film, and current density distribution at the interface of membrane and CL are presented and analyzed for both uniform and non-uniform agglomerate arrangements. The results establish the fact that the distribution of species, water content, electric potential and current density in the CL, i.e. the CL performance, are strongly affected by agglomerate arrangement in the CL. The results of this study can be helpful to improve catalyst layer microstructure, and subsequently its performance. Besides, the presented model can be extended to three dimensions for further investigations of CL microstructure impact.

## 1- Introduction

Numerical models of the CL can be classified in three different categories of thin film models, discrete volume models and agglomerate models. Agglomerate models, which are more accurate and more theoretically detailed relative to other models [1], are based on an assumption that the CL is a matrix of agglomerates which are covered with a thin film of electrolyte.

To the best of author's knowledge, in all simulations carried out by agglomerate models of the CL, the arrangement of agglomerates is considered uniform, while in the reality, due to non-homogenous and anisotropic microstructure of the CL, it is not the case. Only pore-scale numerical methods, such as Lattice Boltzmann Method (LBM) and Pore Network Method (PNM) can capture the non-uniform agglomerates arrangements. The importance of simulating non-uniform agglomerate arrangements becomes more obvious when we realize that manipulating the CL microstructure can significantly enhance its performance and durability [2].

## 2- Numerical Model

The numerical model is based on the LBM with a single relaxation time collision operator (the so-called BGK model) and the popular D2Q9 lattice scheme. To simulate single-phase multi-species gas flow, an active approach which is more accurate relative to passive approach, is adopted [3]. Due to multi scale structure of a PEMFC electrode, multi scale LB simulation is performed by interpolating corresponding density distribution functions of the neighboring zone.

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In the model applied here, the CL is considered as a matrix of cylindrical agglomerates. These agglomerates can be of different sizes and can be unevenly distributed in the CL. Each of the agglomerates can be thought of as a cluster of carbon black particles with platinum catalysts dispersed on their surfaces saturated by an electrolyte material such as Nafion. Moreover, each of these agglomerates is covered by a thin film of electrolyte. Although the penetrated electrolyte (inside of an agglomerate) and the covering electrolyte film (outside of an agglomerate) are connected to each other as a single phase, they are distinguished from each other to clarify agglomerate surface boundary.

Several processes are considered in this model, such as: chemical species penetration from gas channel towards CL pore spaces, dissolution of penetrated oxygen at the pore-electrolyte film interface, diffusion of dissolved oxygen in the electrolyte film towards agglomerate surface, diffusion of dissolved oxygen inside the agglomerate towards reactive sites, transport of proton in the electrolyte covering film., reduction of oxygen on the reactive sites by the electrochemical reaction, back diffusion of produced water at the reactive sites towards the agglomerate surface, back diffusion of produced water from agglomerate surface towards pore-electrolyte film interface and evaporation of water from the pore-electrolyte film interface into pore spaces between agglomerates.

To validate the model, well-known Henry's law model was adopted. The maximum departure of the model results from Henry's law was seen to be 0.25 % which indicates the sufficient accuracy of the model.

A uniform agglomerate arrangement, and a non-uniform arrangement of agglomerates with different sizes are considered for the CL. In the uniform arrangement, which

is the base case, similar columns of cylindrical agglomerates are placed on the membrane. Each column consists of three agglomerates of 2.5 μm radius whose centerline is 7.5 μm away from the centerline of neighboring columns, as shown in Fig. 1. The non-uniform arrangement is obtained by changing the position or radius of some agglomerates of the uniform arrangement.
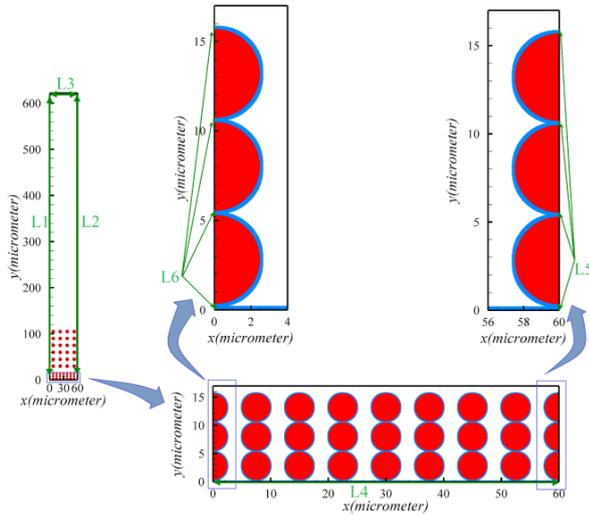


**Figure 1. Computational domain consisting of gas channel, GDL and CL; the agglomerates in the catalyst layer with electrolyte film covering are also shown**

## 3- Results and Discussion

Oxygen mole fraction in the CL and lower portion of GDL are shown in Figures 2(a) and 2(b) at 14.793 ms for two simulations with uniform and non-uniform agglomerate arrangements, respectively. Mole fraction of oxygen is observed to decrease both in the direction of its penetration towards the membrane between the agglomerate columns, and along its bulk transport in the CL. This reduction is due to oxygen consumption in the electrochemical reaction. Fig. 2(b) indicates a non-smooth decrease in oxygen density along the catalyst layer for non-uniform arrangement of agglomerates. This is caused by the disturbed nature of the velocity field, which in turn is due to non-smooth surface of the catalyst layer (composed of non-uniform agglomerate columns). A comparison of Figures 2(a) and 2(b) demonstrates that the (more realistic) non-uniform arrangement of agglomerates can result in a non-homogeneous oxygen distribution. This fact further highlights the importance of implementation of numerical methods, such as the LBM, which can take into account the non-uniformity of agglomerates in the catalyst layer.

Fig. 2(b) also indicates that where agglomerates are closer to each other, such as the fourth and fifth columns from left, oxygen density decreases more severely between the agglomerates. This is so because oxygen penetration through such compact agglomerates occurs at the expense of higher gradients of oxygen density.

The water content of the electrolyte film is shown in Fig. 3. The water content of the electrolyte film remains at its initial value only at the top of columns where it is in contact with the inlet air flow, while elsewhere it increases due to up taking of



(a)



(b)
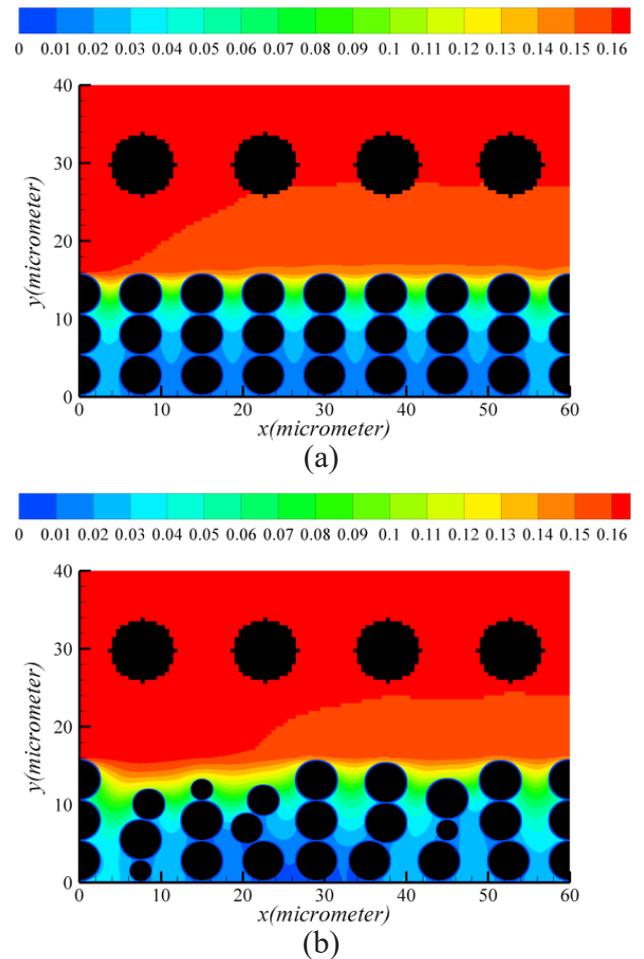
**Figure 2. Computational domain consisting of gas channel, GDL and CL; the agglomerates in the catalyst layer with electrolyte film covering are also shown**

produced water on the surface of agglomerates. A comparison of Figures 3(a) and 3(b) reveals that when the arrangement of agglomerates is non-uniform, the increase of water content from the CL surface down to membrane is non-uniform and even non-monotonic. Fig. 3(b) also illustrates that where the agglomerates are closer to each other, the water content is higher.



(a)
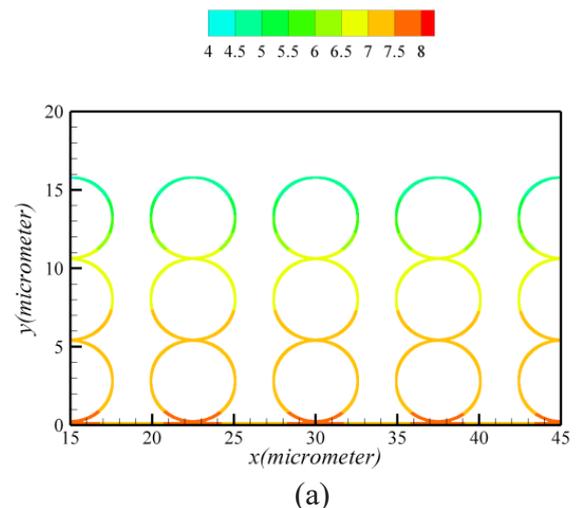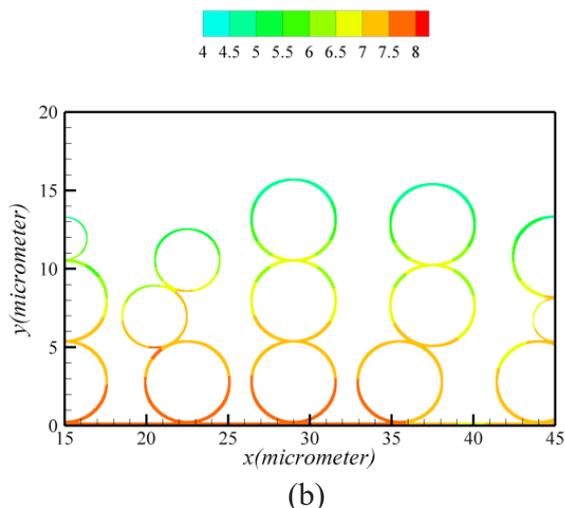
**Figure 3. Local water content distribution in the electrolyte film for two different agglomerate arrangements after 14.793 ms; (a) uniform arrangement, (b) non-uniform arrangement**

## 4- Conclusion

The LBM is adopted in a two-dimensional, pore-scale, single-phase agglomerate model of electrochemically reactive cathode side of a PEMFC. Several processes are taken into account. With the help of this model, the reactive air flow in a cathode electrode is simulated through a multi-scale lattice Boltzmann approach. The oxygen and water vapor distributions in the pore region, the water content and local electrical potential distributions in the electrolyte film and the current density distribution on the interface of CL and membrane are presented for both uniform and non-uniform arrangements of agglomerates. The results demonstrate that distribution of species and current density, and as a result the CL performance, are strongly affected by agglomerate arrangement. Further efforts to improve CL agglomerate arrangements may be precious.

## References

[1] Harvey, D., J. G. Pharoah, and K. Karan, "A comparison of different approaches to modelling the PEMFC catalyst layer", *Journal of Power Sources*, 179 (2008): 209–219.

[2] Khan, M. A., B. Sundén and J. Yuan, "Analysis of multi-phase transport phenomena with catalyst reactions in polymer electrolyte membrane fuel cells – A review", *Journal of Power Sources*, 196 (2011): 7899– 7916.

[3] Chen, S. and G. D. Doolen, "Lattice Boltzmann method for fluid flows", *Annual Review of Fluid Mechanics*, 30 (1998): 329–364.