The Effects of Catalyst Layer Composition onthe Polymer Electrolyte Membrane (PEM) Fuel Cell Performance: (I) Modeling

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***Abstract*— The optimisation problem for the PEM fuel cell catalyst layer was investigated by using the concept of percolation theory. The present work aims to introduce a simple model exploring the effect of catalyst layer composition parameters on the catalyst layer performance. Four design parameters were involved in this study, the Nafion ionomer volume fraction, the pores volume fraction, the catalyst loading and the catalyst layer thickness. Based on the modelling results, it can be concluded that the catalyst layer performance depends highly on the Nafion volume fraction, the pore volume fraction, and the catalyst layer thickness. It was observed that when the Nafion volume fraction approaches to the pore volume fraction, the corresponding catalyst layer thickness is the optimum thickness and the maximum catalyst layer performance was observed. This finding is enhanced by the numerical results, as example, the optimum gemetrical parameters for catalyst layer containing 5.4 mg/cm^2 palladium catalyst loading and 27 µm catalyst thickness, are 0.49 Catalyst Volume Fraction, 0.27 Nafion Volume Fraction, and 0.24 Pores Volume Fraction. Moreover, the catalyst layer thickness is dependent on the catalyst loading; with increase in the catalyst loading the thickness should be increased in order to keep the equality between the pore volume fraction and the Nafion volume fraction.**

Keywords -- PEM Fuel Cells, Catalyst Layer, Percolation Theory, Catalyst Layer Optimization, Renewable and Clean Energy.

I. INTRODUCTION

Catalyst layer (CL) is one the key components for achieving a high performance with reducing costs of a polymer electrolyte membrane (PEM) fuel cell. The structure and elemental distribution of the composition in the catalyst layer determine a fuel cell efficiency, precious catalyst loading and a fuel cell durability [1-5].

Table I. Operating parameters and physical properties of the base-case simulation

Parameter	Symbol	value
Percolation threshold	X^0	0.19
Carbon density	ρ_C	2×10^3 mg/cm^3
Palladium density	ρ_{Pd}	12×10^3 mg/cm^3
Nafion density	ρ_N	1.9×10^3 mg/cm^3
Catalyst layer thickness	L	Variable
Catalyst layer surface area	A	3.14 cm^2
Palladium mass fraction on the supported carbon	β	0.30
Nafion ionomer loadings	X_N	Variable
Catalyst loadings	X_{CAT}	Variable
Average number of the bond neighbours	M	4
Operating temperature	T	300 K
Constant	γ	0
Constant	τ	2
Constant	a	53.7
Constant	b	3.2
Density of an infinite cluster	$P(X_i)$	Calculated
Effective current density in terms of volume fraction	J^{eff}	Unknown
The current density in terms of volume fraction	J	Calculated
Effective area	A^{eff}	Unknown
Pores volume fraction	Y_V	Calculated
Nafion ionomer volume fraction	Y_N	Calculated
Catalyst volume fraction	Y_{CAT}	Calculated
Carbon volume fraction	Y_C	Calculated
Palladium volume fraction	Y_{Pd}	Calculated
Protonic conductivity	σ	Calculated
Effective protonic conductivity	σ^{eff}	Unknown
Catalyst loadings	m_{CAT}	Variable
Palladium loadings	m_{Pd}	Calculated
Carbon loadings	m_C	Calculated
Nafion loadings	m_N	Variable
Catalyst layer current	I	Calculated
Effective Catalyst layer current	I^{eff}	Calculated

The catalyst layer is a heterogeneous porous composite structure of carbon aggregations (electronically conductive), ionomer (transport- ing protons), catalytic nanoparticles to promote the electrochemical anodic hydrogen oxidation and the cathodic oxygen reduction reactions take place (usually platinum or palladium nanoparticles) and pores channels act as a gas transport and distribution paths inside the catalyst layer. Fabrication methods of the PEM fuel cell catalyst layers have attracted a great deal of attention in terms of optimization to satisfy the twin demands of high efficiency and low costs [6]. In CLs, transport paths for protons, electrons, and gases are established by Nafion, carbon, and pore channels, respectively. Inadequate paths can limit the catalyst layer performance. For these reasons, a fundamental understanding of the structures of CLs and their relationships with PEM fuel cell performance is urgently required.

Percolation theory is used to describe a variety of natural physical processes, which have been discussed in detail by Stauffer and Aharony [7] and Sahimi [8]. In two-dimensional percolation, either exact values or precise estimates are known for the critical thresholds and other related coefficients and exponents [7-12]. However, three-dimensional lattices are relevant for most natural processes. The most common of these are the simple cubic, (sc), the face-centred-cubic, (fcc), and the body centred-cubic bcc lattices. The percolation thresholds for these lattices are not known exactly and the estimates that have been determined for the latter two lattices are much less precise than the values that have been found for typical two dimensional systems. A general introduction to percolation theory is presented in references [7-12]. Authors who have appealed to percolation theory to explain or predict electrode properties in fuel cells have generally either used advanced simulation approaches that consider particles of specific shape [13], or have just noted that there is an optimum composition and that this is consistent with some connectivity criterion [14]. The present work aims to introduce a simple model exploring the effect of catalyst layer composition parameters such as Nafion volume fraction, porosity, or void volume fraction, catalyst (Pd/C) loading, and the catalyst layer thickness on the PEM fuel cell catalyst layer performance using the principles of percolation theory.

II. PERCOLATION MODEL

The microstructural studies [12,15] of the catalyst layer suggested single agglomerates as the structural unit of the PEM fuel cell catalyst layer. These agglomerates comprise palladium supported carbon, mixed with Nafion ionomer, gas pores and water filled pores. The expansion of the reaction zone depends on the composition, surface boundary conditions, and the radii of the agglomerates. Due to, these dependencies the contributions of single agglomerates to the catalyst layer performances depend on their position within the macroscopic catalyst layer. These effects are conventionally described by the so called effectiveness factor [16]. As indicated in Fig.1, the effective Pd catalyst particles in the cluster are those that are in contact with the Nafion ionomer and the carbon particles, i.e. the particles which have electronic channels via the carbon particles to current collector and protonic channels via Nafion ionomer particles to the Nafion membrane. Consequently, the macroscopic transport of protons, reactant gases and the electrons from and to the highly dispersed palladium sites determines the overall electrode performance. The system is thin and nearly two-dimensional (the nearly is important). If it were truly two-dimensional, the applicable continuum percolation threshold (X^{O}) would be around 0.45 [16] - that is, connectivity of both phases would only be achieved between the Nafion volume fractions X_N = 0.45 and X_N = 0.55. The highest probability of having good connectivity of both phases is at X_N = 0.5. This is a very tenuous criterion; since real particulate systems with round particles do not conform exactly to the truly random continuum percolation geometry (positive curvature predominates, whereas a random continuum has zero net curvature). It is quite likely that the real threshold for our system is greater than X_N = 0.5; under these circumstances, continuity of both phases can never be achieved in two dimensions. A simple example of such a system is the two-dimensional square lattice, which has X^{O} (site) of 0.59 - random occupancy of such a lattice by two types of entity never leads to site percolation of both entities. By introducing the catalyst layer thickness, the two-dimensional thresholds are relaxed and continuity of both phases can be achieved over a certain range around X^{O} = 0.5. The optimum will always be X^{O} = 0.5 if the two phases are geometrically similar. There are theorems that relate the percolation probability in a finite-size system to the composition and the size[s] of the finite dimension[s] [11]. In our system the two phases are not similar – the solid polymer electrolyte (SPE), being originally a liquid, is more able to adopt both positive and negative curvatures; thus we would predict that the optimum composition would be displaced slightly towards lower X_N - that is, less SPE and more conductor are required to give them equal connectivity. The face centre cubic lattice is a common and closest approximation for any densely packed structures. Therefore, it can be chosen as the best approximation to describe the catalyst layer in terms of the percolation theory. The catalyst layer operation involves effective transport of gas, water, and protons due to the electrochemical reactions. The effective parameters that steer the interplay of these processes are proton conductivity and the exchange current density. These parameters incorporate information about the composition and the pore structure. In order to be able to rationalise the major structure effects, simplified expressions based on the percolation theory are employed.

III. CATALYST LAYER COMPOSITION PARAMETERS

PEM fuel cell catalyst layer can be designed in in terms of mass fractions of the consistent materials [12]. Therefore, it would, be very useful to link the performance directly to the mass fractions of the catalyst layer components.

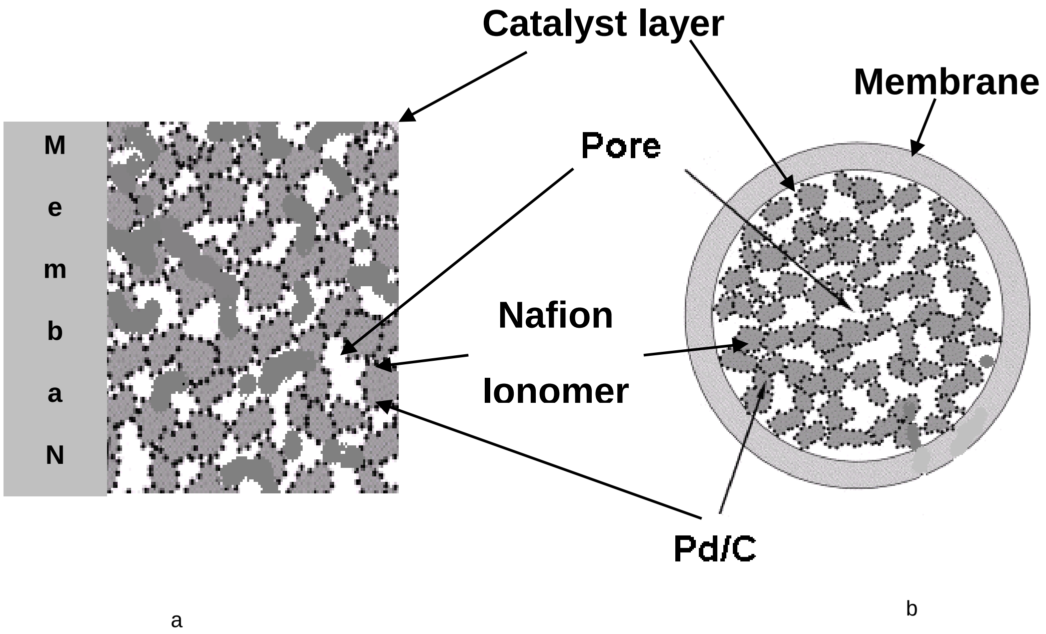

Figure 1. Structural diagram of the Membrane Electrode Assembly (MEA), depicting the basic catalyst layer structure: (a) cross sectional sketch and (b) surface sketch.

Palladium, carbon, and the Nafion ionomer mass fractionsX_{Pd}, X_C , X_{Pd} , are determined from the mass percentage of palladium, carbon, and Nafion respectively using the following relations

$$X_N = \frac{m_N}{m_{CL}} \tag{1}$$

$$X_{Pd} = \frac{m_{Pd}}{m_{CL}} \tag{2}$$

$$X_C = \frac{m_C}{m_{CL}} \tag{3}$$

Where m_N ,m_C,m_{Pd} are the Nafion, Carbon, Palladiun, masses per unit area in units of mg/ cm^2 , and m_{CL} is the total mass of the catalyst layer per unit area in units of mg/ cm^2.

$$m_{CL} = m_N + m_C + m_{Pd} \tag{4}$$

By including the catalyst layer thickness L the effective catalyst layer properties will be specified by the volume fractions of Nafion ionomer X_V, palladium Y_{Pd} and carbon Y_C.
In addition, the pores volume fraction Y_V can be evaluated

$$Y_N = \frac{m_{Pd} X_N}{\rho_N \beta L (1 - X_N)} \tag{5}$$

$$Y_{Pd} = \frac{m_{Pd}}{\rho_N L} \tag{6}$$

$$Y_C = \frac{m_{Pd}(1 - \beta)}{\rho_N \beta L} \tag{7}$$

and the pores volume fraction $Y_V$is:

$$Y_V = 1 - Y_N - Y_{Pd} - Y_C \tag{8}$$

Where, ρ_{Pd} , ρ_C , ρ_N are the densities of palladium, carbon, and Nafion respectively, and β is the palladium mass fraction on the supported carbon black (in this study $\beta = 0.3$). The relation between these volume fractions and the effective catalyst layer parameters such as; the effective proton conductivity σ^{eff} , the effective gas diffusion coefficient D^{eff}, and the effective exchange current density J^{eff} can be established in terms of percolation threshold X^0.

IV. CATALYST LAYER PERFORMANCE PARAMETERS

PEM fuel cell catalyst layer performance can be evaluated by means of the effective protonic conductivity, effective gas diffusivity, and the effective current density.
These parameters are evaluated using the catalyst layer structure parameters.

A. Effective Protonic Conductivity

The effective proton conductivity is determined by the volume fraction of the ionomer electrolyte, expressed as [8-12]:

$$\sigma^{eff} = \sigma\left(\frac{Y_N - X^0}{1 - X^0}\right)^{\tau} \Theta(Y_N - X^0) \tag{9}$$

Where σ is the bulk proton conductivity; τ is the critical exponent (which is universal, i.e., does not depends on the catalyst layer structure but depends on the system dimensionality, (In three dimensions) $\tau \approx 2$, Θ is the Heaviside-step-function, which accounts for zero conductivity below the percolation threshold, X^0 , and X_N is the Nafion volume fraction [12, 17-19];

$$\Theta(Y_N - X^0) = \begin{cases} 0 \ \ at\ Y_N < X^0 \\ 1 \ \ at\ Y_N \geq X^0 \end{cases} \tag{10}$$

The percolation threshold X^0 depends on the geometry of the lattice. If the pore space is highly coordinated, as in a densely packed structure where a single pore has 12 nearest neighbour pores, X^0 is small (in this case = 0.19 for the site-percolation).

B. Effective Diffusion Coefficient.

At the cathode catalyst layer, the diffusion of oxygen in the catalyst layer is more complex. Oxygen in the gas phase transports to the reaction sites by diffusion through a percolation cluster of the pores and by residual diffusion of dissolved oxygen through the catalyst layer primary pores The bulk diffusion coefficient in the secondary gas pores is larger than in the primary pores by a factor of [19,20]. The latter is considered by a residual diffusivity, and the effective diffusion coefficient can be expressed as, [17,19].

$$D_{O_2}^{eff} = \frac{D_{O_2}}{(1 - X^0)^{\tau} + d}(Y_V - X^0)^{\tau}\Theta(Y_V - X^0 + d) \tag{11}$$

Where D_{O_2} , is the bulk oxygen diffusion coefficient. The coefficient d accounts for the diffusivity due to diffusion through primary pore. In this study the primary pores were assumed to be blocked by the Nafion ionomer "which means that $d = 0$ ". The critical exponent of diffusion,τ is universal for standard lattice percolation.[7] Values of $\tau \approx 2$ are suggested in three dimensions by theoretical studies [9]. However, we are aware that for specific structures, including fibrous structures [11] and for distinct types of continuum percolation models, e.g., the so-called Swiss cheese model [12], different critical exponents could be found. However, in view of the yet unresolved structural picture of cathode catalyst layer we believe that $\tau \approx 2$ is a reasonable choice, because it apparently can be used for a broad class of random heterogeneous structures.

C. Effective Current Density

The electrochemical active catalyst layer area can be defined as the specific area of the palladium / Nafion interface which is accessible for protons, as well as for the gas reactant (oxygen at the cathode and hydrogen at the anode), and it depends on the volume fractions of the catalyst layer compositions. Therefore, the effective catalyst layer area A^{eff} can be approximated as [19]:

$$A^{eff} = AP(Y_N)P(Y_{Cat})\left\{\left[1 - \left(1 - P(Y_V)\right)^M\right](1 - \gamma) + \gamma\right\} \tag{12}$$

where the term in the square brackets represents that there is at least one pore as a neighbor of the bond drawn between the Nafion ionomer and the catalyst particle, the factor $P(Y_N)P(Y_{Cat})$ represents the probability of an interface between the catalyst and the Nafion ionomer with both of them connected to their corresponding infinite clusters, and $P(Y_i)$ is the density of an infinite cluster of a percolating component, which is determined by:

$$P(Y) = \frac{Y}{[1 + \exp(-a(Y - X^0))]^b} \tag{13}$$

Where a = 53.7, b= 3.2, as considered in reference [19].

The PEM fuel cell output current is:

$$I = I^{eff} \tag{14}$$

Where, I^{eff} is the effective output current. Therefore, the current density j in terms of volume fraction of the catalyst layer composition can be approximated as:

$$j = j^{eff}\frac{A^{eff}}{A} \tag{15}$$

or, according to equation (12), $j$can be expressed as:

$$j = j^{eff}P(Y)P(Y_{cat})\left\{\left[1 - \left(1 - P(Y_V)\right)^M\right](1 - \gamma) + \gamma\right\} \tag{16}$$

where j^{eff} is the volume current density of the three-phase composite zone, and M is the average number of the bond neighbors (for fcc M = 4). The parameter γ is the residual diffusivity parameter. In the present work γ was taken to be zero [17].

D. Code Description

The operating parameters and physical properties of the base-case simulation are listed in Table I. The Nafion volume fraction, pores volume fraction catalyst loading and catalyst layer thickness are related by the equations (5 – 8). In the following optimisation the effect of composition parameters on the sensor performance investigated. A simple Matlab computer program was written and used in this study.

V. RESULTS AND DISCUSSION

A. *Effect of Nafion Volume Fraction*

Fig. 2, shows the dependence of the proton conductivity on the Nafion ionomer volume fractions for a catalyst layer containing 5.4 mg/cm^2 catalyst loading and its thickness of 25.0 µm.

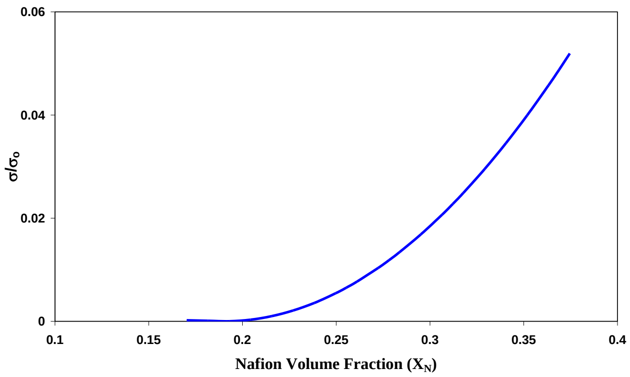

Figure 2. The dependence of the proton conductivity on the Nafion ionomer volume fractions in PEM fuel cell catalyst layer

From the figure it can be seen that above the percolation threshold the gradually increasing in Nafion volume fraction (with constant thickness) leads to increase in the protonic conductivity of hydrated catalyst layer. However, the catalyst layer performance is very sensitive to the Nafion volume fraction. Fig. 3, shows, the effect Nafion volume fractions on the catalyst layer performance for a catalyst layer.

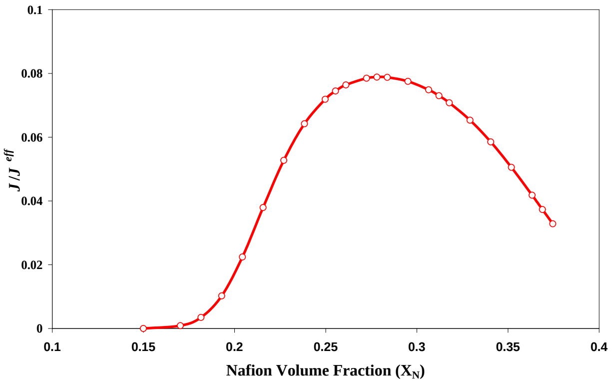

Figure 3. The effect Nafion volume fractions on the catalyst layer performance in the PEM fuel cell catalyst layer.

As the Nafion volume fraction increases the catalyst layer performance increases and gradually levels off to a high value. Further increases of the Nafion volume fraction can result in a decreased current density, especially for the cases of thinner catalyst layers and high catalyst loadings. These results can be explained as follows. The Nafion volume fraction is closely related to the proton transport capability of the catalyst layer. When the Nafion volume fraction is low, the poor proton transport is a critical factor to the catalyst layer performance and, consequently, increasing the Nafion volume fraction can improve the catalyst layer performance. On other hand, too high a Nafion ionomer volume fraction causes reduction of catalyst layer performance due to blocking of the catalyst sites, blocking of the catalyst layer pores, reduction of gas permeability and increased mass transfer overpotential. Hence, the icrease in Nafion volume fraction will results in decrease in the reactant gas diffusion and hence fast decline of the current density will be observed. Therefore, based on this model, for catalyst layer containing 5.4 mg/cm^2 catalyst loading and 25 µm catalyst layer thickness the optimum Nafion ionomer volume fraction was 0.278.

The model has been compared with the experimental results [21]. The experimental conditions were used as input data, and the output results are presented in Fig. 4. The results showed highly correlation between the experimental and numerical results for the optimum Nafion ionomer volume fractions.

It can be seen that numerically the optimum Nafion ionomer volume fraction was in the range 0.29 to 0.35, while from experimental point of view, the optimum Nafion ionomer volume fraction was in the range of 0.31 to 0.35. It should also be noted that Nafion is assumed to be fully humidified in the present simulation. If Nafion is drier and hence the proton conductivity is lower, the Nafion volume fraction will have more pronounced effects on the catalyst layer performance.

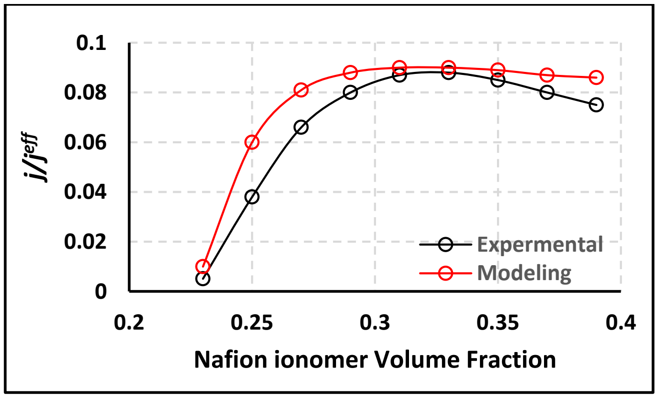

Figure 4.Comparisons between the experimental data [21], and the numerical data obtained by this model at maximum catalyst layer performance for a catalyst layer containing 5.4 mg/cm^2 catalyst loading and its thickness of 25.0 µm.

B. *Effect of Pores Volume Fraction*

The optimum catalyst layer pore volume fraction for the electrodes of 5.4 mg/cm^2 catalyst loading and catalyst layer thickness of 25 µm was evaluated numerically and the results were presented in Fig.5. The results showed that for porevolume fractions less than 0.246 the catalyst layer performance drops rapidly.

As the pore volume fractions and permeability decrease, reactant transport rate by diffusion and advection within the catalyst layer drops sharply. According to this model, for catalyst layer containing 5.4 mg/cm^2 catalyst loading and 25 µm catalyst layer thickness the optimum Nafion ionomer volume fraction was 0.278. In addition, the inverse relation between the voids volume fraction and the protonic conductivity of catalyst layer was observed at constant thickness.

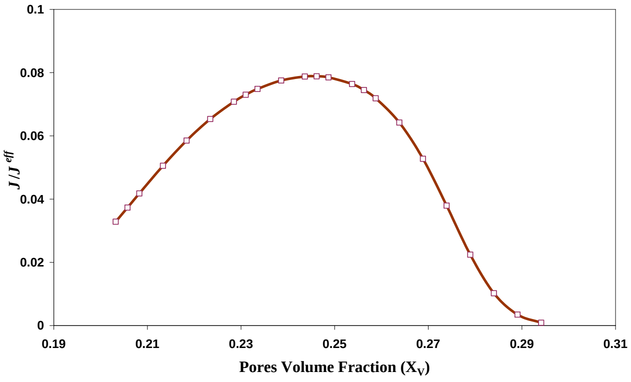

Figure 5.Pores volume fraction effects on the catalyst layer performance

The model has been tested using the experimental information which presented ref. [21], and the results are presented in Table II. It can be seen that numerically the optimum pores volume fraction was in the range 0.305 to 0.307, while from experimental point of view, the optimum pores volume fraction was in the range of 0.272 ± 0.005 to 0.275 ± 0.003.

C. Effect of Catalyst Layer Thickness

The catalyst layer thickness also plays a role in the relations between the catalyst layer composition volume fractions and its performance. Fig. 6, illustrates the catalyst layer thickness effects on the MEA performance. The result showed that the optimum catalyst layer thickness is 27.5 µm. At lower thickness values, the catalyst layer performance is low, i.e. there is mass transfer problem. As the catalyst layer thickness increased, the MEA performance increase. Further increase in the catalyst layer thickness is results in a slowly decreased MEA performance. Therefore, the catalyst layer thickness must be optimised to control the porosity, the Nafion ionomer and the catalyst volume fractions. In similar way, the model has been tested using the experimental information [21], and the results are presented in Table II. The results showed that when the Nafion ionomer and the pore volume fraction are very close to each other the corresponding catalyst layer thickness is the optimum one. Therefore, it can be expected that the maximum MEA performance can be obtained when the volume fractions of the Nafion ionomer and the pores are equal.

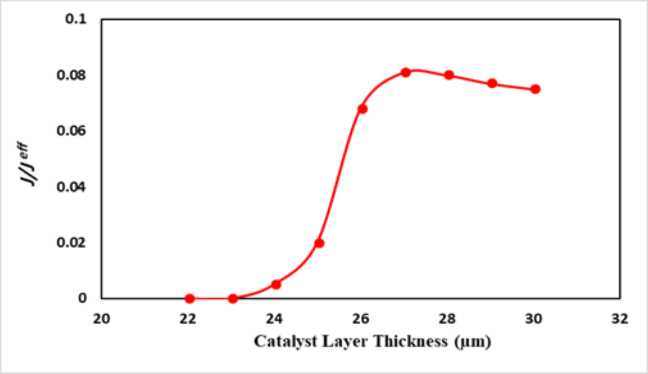

Figure 6. Catalyst layer thickness effects on the catalyst layer performance.

Table II. Comparisons between the experimental data obtained in ref. [21], and the numerical data obtained by this model at maximum cell performance and the same conditions

Test No.		**Catalyst layer Weight (mg)**	**Nafion Loading wt %**	**Catalyst layer (µm)**	**Catalyst layer Composition Volume Fraction**		
					Catalyst	**Nafion**	**Pores**
1	Experimental	10.47	25	16.1	0.454	0.273	0.274
	Numerical	10.47	30	18.6	0.392	0.306	0.305
2	Experimental	13.7	25	21.1	0.453	0.272	0.275
	Numerical	14.7	30	24.5	0.391	0.302	0.307
3	Experimental	17.2	25	25	0.455	0.273	0.272
	Numerical	18.3	30	32.6	0.388	0.306	0.306

In the present study, showed that, the optimum gemetrical parameters for catalyst layer containing 5.4 mg/cm^2 palladium catalyst loading and 27 µm catalyst thickness, are 0.49 Catalyst Volume Fraction, 0.27 Nafion Volume Fraction, and 0.24 Pores Volume Fraction.

VI. CONCLUSION

A simple numerical model is developed using percolation theory principles to relate the catalyst layer performance with the catalyst layer composition. Four catalyst layer parameters were included: The Nafion ionomer volume fraction, the porosity volume fraction, the catalyst layer thickness and the catalyst (Pd/C) loading. Based on the modelling results, it can be concluded that the catalyst layer performance depends highly on the Nafion volume fraction, the pore volume fraction, the catalyst loading and the catalyst layer thickness. When the Nafion volume fraction approaches to the pore volume fraction, the corresponding catalyst layer thickness is the optimum catalyst layer thickness and the maximum catalyst layer performance was observed. Moreover, with increase in the catalyst loading the catalyst layer thickness should be increased in order to keep the equality between the pore volume fraction and the Nafion volume fraction. The accuracy of the model is highly dependent on the mathematical and statistical description of the phenomena in the catalyst layer. Further theoretical studies should be done in order to develop the model.

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