

Analysis of Carbon Particle Overlap Effects on Oxygen Reactive Transport in Catalyst Layers of Proton Exchange Membrane Fuel Cells

Ruiyuan Zhang¹, Li Chen^{1*}, Wen-Quan Tao¹

¹ Key Laboratory of Thermo-Fluid Science and Engineering of MOE, School of Energy and Power Engineering, Xi'an Jiaotong University, Xi'an, Shaanxi, 710049, China (*Corresponding Author)

ABSTRACT

Reducing the Pt loading, especially on the cathode side, is very important for reducing the cost of proton exchange membrane fuel cells. However, lowering the Pt loading causes high voltage losses, but the underlying mechanism has not been fully understood. In this study, a new analytical catalyst layer model is established to study the oxygen reactive transport process, in which effect of carbon particle overlap is considered. Results from the analytical model show that carbon particle overlap leads to increase of ionomer thickness, reduction of ionomer specific surface area, and finally increase of oxygen transport resistance. By considering carbon particle overlap, an increased ~ 100 nm effective ionomer thickness is observed, and creates ~ 1500 s m⁻¹ ionomer resistance. The order of magnitude of such ionomer resistance is equivalent to that of the Pt surface resistance measured in previous experimental studies, but there is no clarified source of this Pt surface resistance. As a result, it is reasonable to consider that carbon particle overlap may cause one of the sources of the oxygen transport resistance, which provides new insights for further alleviating the oxygen transport resistance under low Pt loading.

Keywords: catalyst layer, low Pt loading, carbon particle overlap, ionomer film, oxygen transport resistance

1. INTRODUCTION

Proton exchange membrane fuel cell (PEMFC) is one of the most promising eco-friendly devices. Expensive platinum (Pt) is widely used as catalysts in the membrane electrode assembly (MEA), and accounts for the most significant cost in a fuel cell stack [1]. The Pt loading (Pt amount per unit surface area of MEA)

should be reduced to achieve low cost of PEMFC for commercial applications.

However, with the decrease of Pt loading in MEAs, additional voltage loss occurs especially in high current density region [2-4]. This voltage loss is mainly due to the oxygen transport resistance in the catalyst layer [3, 5, 6]. A lot of work has been done to study the oxygen transport resistance under low Pt loading [2, 5-10], but the source of oxygen transport resistance is still not very clear. Greszler et al. [5] changed the Pt loading over a wide range from 0.4 mg cm⁻² to 0.03 mg cm⁻², and found that the oxygen transport resistance is inversely proportional to the surface area of Pt particles. The scale coefficient is calculated as 1200 s m⁻¹, which is mathematically similar to a resistive film on the Pt surface. The film-like resistance is equivalent to a ~ 35 nm bulk ionomer, but in reality the typical ionomer thickness is only 4-10 nm in the catalyst layer based on the experimental observation. In order to clarify the origin of this film-like resistance, Kudo et al. [6] studied the oxygen transport resistance of the ionomer thin film in a range of 20-100 nm. They calculated the film-like resistance and found it is around 1800 s m⁻¹, equivalent to 30-70 nm of the ionomer film. Similarly, Ono et al. [9, 11] reported in their study that the film-like resistance is around 1240 s m⁻¹. Interfacial resistance at the pore/ionomer interface is an appealing hypothesis [12-14]. However, such hypothesis has not been completely confirmed by the experimental results. Before more solid verifications, it is unwise to consider that the interfacial resistance is the only source of the film-like resistance.

In this paper, a new analytical catalyst layer model is established to study the oxygen reactive transport process. Based on the actual microscopic morphology of the catalyst layers, carbon particles can overlap each

other and have a specific overlap ratio [2,3]. Due to the overlap of carbon particles, the thickness of ionomer film on carbon surface is expected to increase, which can partially clarify the origin of oxygen transport resistance under low Pt loading.

2. MODEL CONSTRUCTION

2.1 Catalyst layer structure modelling

In this study, a catalyst layer model considering the carbon particle overlap is constructed. Based on a single carbon particle, the effects of carbon particle overlap on ionomer distribution on the surface of the carbon particle are studied in detail.

Fig. 1(a) illustrates a single Pt/C particle without overlap, where red, black and green parts represent Pt particles, carbon particle and ionomer. Assuming spherical shape of this ionomer-coated Pt/C particle, the following relationships can be obtained,

$$V_{\text{total}} = \frac{4}{3} \pi (r_c + \delta_N)^3 \quad (1)$$

$$V_c = \frac{4}{3} \pi r_c^3 \quad (2)$$

$$V_{\text{Pt}} = n_{\text{Pt}} \frac{4}{3} \pi r_{\text{Pt}}^3 \quad (3)$$

$$V_N = V_{\text{total}} - V_c - V_{\text{Pt}} \quad (4)$$

where V_{total} represents the volume of entire structure, and V_c , V_{Pt} and V_N represent the volume of carbon particle, Pt particles and ionomer respectively. r_c and r_{Pt} are the radius of carbon and Pt particles. δ_N is the ionomer thickness. n_{Pt} is the number of Pt particles on carbon particle surface. If a certain I/C (ionomer to carbon weight ratio) is given, V_N can be calculated as,

$$V_N = \frac{I/C}{\rho_N/\rho_c} V_c \quad (5)$$

Besides, n_{Pt} is obtained by a certain Pt/C wt% (weight ratio of Pt to Pt/C catalysts),

$$V_{\text{Pt}} = \frac{\text{wt}\%}{1 - \text{wt}\%} \frac{\rho_c}{\rho_{\text{Pt}}} V_c \quad (6)$$

$$n_{\text{Pt}} = \frac{V_{\text{Pt}}}{\frac{4}{3} \pi r_{\text{Pt}}^3} = \frac{\text{wt}\%}{1 - \text{wt}\%} \frac{\rho_c}{\rho_{\text{Pt}}} \left(\frac{r_c}{r_{\text{Pt}}} \right)^3 \quad (7)$$

where ρ_c and ρ_{Pt} are densities of carbon and Pt, respectively. r_{Pt} can be calculated if the electrochemical specific surface area (ECSA) of Pt is given,

$$r_{\text{Pt}} = \frac{3}{\rho_{\text{Pt}} a_{\text{ECSA}}} \quad (8)$$

Carbon particle overlap has not been considered in the above model derivation, so the carbon particle coated with ionomer is a regular sphere. When the carbon particle overlap is further considered, the surface area of carbon will decrease and the ionomer distribution space will be compressed. Fig. 1(b) shows the structure of Pt/C particles after considering the carbon particle overlap. It can be seen that the ionomer thickness will increase since the total volume of ionomer remains unchanged (the orange part in Fig. 1(b)). In this study, k is defined as the number of carbon particles that overlap with the studied one, and δ is defined as the radial overlap depth of carbon particles. Given a certain k and δ , the expressions of V_{total} and V_N will be accordingly modified based on volume conservation

$$V_{\text{total}} = \frac{4}{3} \pi (r_c + \delta_N)^3 - k \times \pi (\delta + \delta_N)^2 \left(r_c + \delta_N - \frac{\delta + \delta_N}{3} \right) \quad (9)$$

$$V_c = \frac{4}{3} \pi r_c^3 - k \times \pi \delta^2 \left(r_c - \frac{\delta}{3} \right) \quad (10)$$

Besides, V_{Pt} still follows Eq. (3) since Pt particles do not overlap and V_N is unchanged due to volume conservation.

$$V_N = V_{\text{total}} - V_c - V_{\text{Pt}} = \frac{I/C}{\rho_N/\rho_c} \times \frac{4}{3} \pi r_c^3 \quad (11)$$

Therefore, substituting Eqs. (5), (9) and (10) into Eq. (11), δ_N after carbon particle overlap can be determined

$$A\delta_N^3 + B\delta_N^2 + C\delta_N + D = 0 \quad (12)$$

where A , B , C and D are constants for the above three-order equation,

$$A = \frac{4}{3} \pi - \frac{2}{3} \pi \times k$$

$$B = 4\pi r_c - \frac{4}{3} \pi \delta^2 \times k + \left(\frac{\delta}{3} \pi - \pi r_c \right) \times k \quad (13)$$

$$C = 4\pi r_c^2 - \frac{2}{3} \pi \delta^2 \times k + 2\delta \left(\frac{\delta}{3} \pi - \pi r_c \right) \times k$$

$$D = -\frac{4}{3} \pi \frac{\text{wt}\%}{1 - \text{wt}\%} \frac{\rho_c}{\rho_{\text{Pt}}} r_c^3 - \frac{4}{3} \pi r_c^3 \frac{\rho_c}{\rho_i} \times I/C$$

Eqs. (12)-(13) can be analytically solved. Three solutions can be obtained, while only one real root among the three solutions is the feasible solution.

Then, based on the calculated δ_N , the surface area of carbon and ionomer film can be obtained,

$$S_c = 4\pi r_c^2 - 2\pi r_c \delta \times k \quad (14)$$

$$S_N = 4\pi (r_c + \delta_N)^2 - 2\pi (r_c + \delta_N) (\delta + \delta_N) \times k \quad (15)$$

the specific surface area of ionomer film can be calculated by

$$a_N = \frac{S_N}{V_{\text{total}}/(1 - \varepsilon_p)} \quad (16)$$

where ε_p is the porosity of catalyst layer.

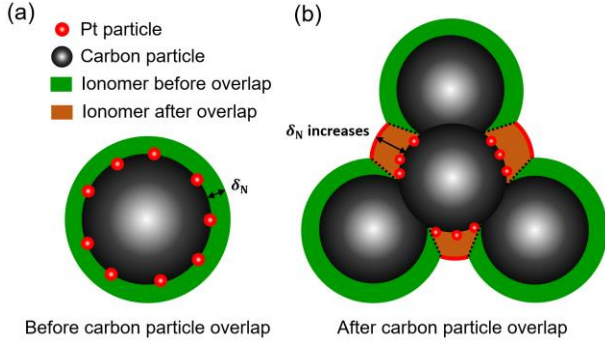


Fig. 1. Schematic of the structure of Pt/C particles before and after carbon particle overlap.

2.2 Local transport process modelling

The local transport process refers to the transport of oxygen in the ionomer film near the Pt particles. This is a very complex transport process. First, driven by the concentration difference, oxygen needs to dissolve from the pores into the ionomer. The dissolution process follows Henry's law,

$$C_{O_2}^N = \frac{RT}{H_N} C_{O_2}^g \quad (17)$$

where H_N is the Henry's constant of oxygen in ionomer. Eq. (17) depicts the equilibrium dissolution of oxygen in ionomer.

Second, the oxygen flux through ionomer thin film is described by Fick's,

$$N_{O_2} = \frac{D_N}{\delta_N} (C_{O_2}^N - C_{O_2}^{Pt}) \quad (18)$$

where D_N is oxygen diffusivity in ionomer and $C_{O_2}^{Pt}$ is the oxygen concentration on Pt particle surface. Note that Eq. (18) follows two assumptions. The first is that the ionomer thickness should be thin enough compared to the carbon radius. The second is that Pt particles completely cover the carbon surface so that ORR homogeneously occurs on carbon surface. However, in fact, Pt particles are discretely distributed on carbon surface especially at a relatively low Pt/C wt%, and thus the diffusion length of oxygen inside ionomer film may not be strictly identical with δ_N . Here, an effective ionomer film thickness δ_N^{eff} is defined to represent the actual diffusion path length of oxygen in ionomer. Therefore Eq. (18) can be rewritten as

$$N_{O_2} = \frac{D_N}{\delta_N + \delta^*} (C_{O_2}^N - C_{O_2}^{Pt}) = \frac{D_N}{\delta_N^{\text{eff}}} (C_{O_2}^N - C_{O_2}^{Pt}) \quad (19)$$

where δ^* indicates the difference between δ_N and δ_N^{eff} , which should always be greater than or equal to zero. δ_N^{eff} is a synthetic parameter considering the structure effects on oxygen diffusion length, and theoretically if a proper δ_N^{eff} is defined, Eq. (19) can accurately evaluate the oxygen transport resistance in ionomer,

$$\Omega_N = \frac{\delta_N^{\text{eff}}}{D_N} \quad (20)$$

where Ω_N is defined as the oxygen transport resistance in ionomer. If further considering the Henry's law at pore-ionomer interface, the oxygen local transport resistance in ionomer film can be obtained,

$$\Omega_{\text{local},N} = \frac{\delta_N^{\text{eff}}}{D_N} = \frac{\delta_N^{\text{eff}}}{P_N} \quad (21)$$

where P_N is the oxygen permeability coefficient in ionomer film.

Third, ORR takes place on Pt surface, which obeys the Butler-Volmer type expression,

$$N_{O_2} = \frac{1}{4F} i_0^{\text{ref}} \frac{C_{O_2}^{Pt}}{C_{O_2,\text{ref}}^{\text{Pt}}} \left[\exp\left(-\frac{\alpha_c F}{RT} \eta\right) - \exp\left(\frac{(1-\alpha_c) F}{RT} \eta\right) \right] \quad (22)$$

$$= k_{\text{elec}} C_{O_2}^{\text{Pt}}$$

where F is Faraday constant. i_0^{ref} is reference current density. $C_{O_2,\text{ref}}^{\text{Pt}}$ is reference oxygen concentration. α_c is charge transfer coefficient. R is ideal gas constant. T is temperature. η denotes overpotential. k_{elec} is the electrochemical reaction rate constant.

Combining Eqs. (19)-(22), the following formula can be obtained,

$$\frac{C_{O_2}^g - C_{O_2}^{\text{Pt}}}{\Omega_{\text{local},N}} = k_{\text{elec}} C_{O_2}^{\text{Pt}} \quad (23)$$

and thus $C_{O_2}^{\text{Pt}}$ can be calculated as,

$$C_{O_2}^{\text{Pt}} = \frac{C_{O_2}^g}{1 + k_{\text{elec}} \Omega_{\text{local},N}} \quad (24)$$

Therefore, the local reaction rate of oxygen can be obtained using $C_{O_2}^{\text{Pt}}$,

$$N_{O_2} = k_{\text{elec}} C_{O_2}^{\text{Pt}} = \frac{k_{\text{elec}}}{1 + k_{\text{elec}} \Omega_{\text{local},N}} C_{O_2}^g \quad (25)$$

Eq. (25) can be easily rewritten by substituting $1/k_{\text{elec}}$ as $\Omega_{\text{local},Pt}$,

$$N_{O_2} = (\Omega_{\text{local},Pt} + \Omega_{\text{local},N})^{-1} C_{O_2}^g = \Omega_{\text{local}}^{-1} C_{O_2}^g \quad (26)$$

where Ω_{local} is the sum of $\Omega_{\text{local,N}}$ and $\Omega_{\text{local,Pt}}$, denoting the total local transport resistance from pores to Pt. At limiting current density, $\Omega_{\text{local,Pt}}$ should be zero. Then,

3. RESULTS AND DISCUSSION

3.1 Effects of carbon particle overlap on Pt/C particle structure

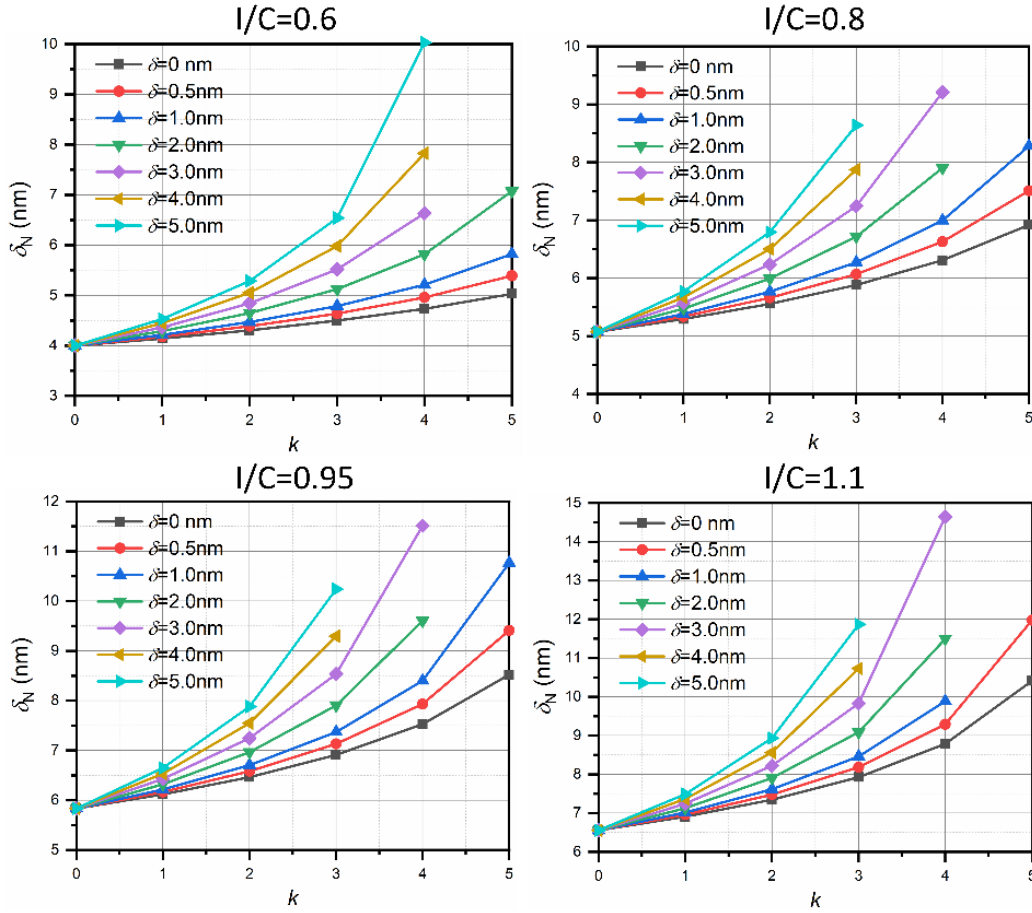


Fig. 2. Variations of δ_N under different k , δ and I/C .

the oxygen reaction rate per unit volume of catalyst layer, defined as ψ , can be calculated by multiplying N_{O_2} and a_N ,

$$\psi = N_{\text{O}_2} a_N = \frac{a_N}{\Omega_{\text{local}}} C_{\text{O}_2}^g \quad (27)$$

Here, $a_N/\Omega_{\text{local}}$ is defined as the effective local reaction rate constant k_{elec}^* , and thus Eq. (27) can be obtained,

$$\psi = k_{\text{elec}}^* C_{\text{O}_2}^g \quad (28)$$

Further, if it is assumed that oxygen is transported very fast in the pores of the catalyst layer, the consumption rate of oxygen is equal everywhere in the whole catalyst layer, and the local transport resistance of oxygen can be obtained by Eq. (29),

$$R_{\text{local}} = (\psi \delta_{\text{CL}})^{-1} \quad (29)$$

where δ_{CL} is the thickness of the entire catalyst layer.

Fig. 2 shows the effects of carbon particle overlap on ionomer thickness δ_N . The range of k is 0-5, and that of δ is 0-5 nm. Four different values of I/C are studied, including 0.6, 0.8, 0.95 and 1.0. Other parameters used in this study are listed in Table 1. Firstly, it can be seen that under each group of I/C , δ_N will increase with the increase of k and δ . For example, at I/C of 0.6, when the carbon particle overlap does not occur, δ_N is only about 4 nm. However, as k rises to 4 and δ rises to 5 nm, the resulted δ_N even exceeds 10 nm. Such result indicates that the carbon particle overlap can increase the ionomer thickness several times. Secondly, the ionomer content will also affect the δ_N . For example, when k equals 4 and δ equals 3nm, it can be seen that at I/C of 0.6, 0.8, 0.95 and 1.0, the δ_N is about 6.63 nm, 9.21 nm, 11.51 nm and 14.64 nm, respectively. Compared to δ_N without carbon particle overlap, the corresponding increase rates of δ_N are 65.83%, 81.45%, 97.38% and 123.36%, respectively. In other words, a higher I/C will cause more significant influence of carbon particle

overlap on ionomer thickness. This is because with the increase of ionomer volume, the ionomer film is forced to thicken along the radial direction of carbon particle, and the increase of carbon particle overlap leads to thicker ionomer.

Table 1. Critical parameters used in model construction [6].

Variable	Symbol	Value
Radius of carbon	r_c	25 nm
Catalyst layer thickness	δ_{CL}	10 μm
Porosity	ε_p	0.4
Henry's constant	H_N	$2.21 \times 10^4 \text{ Pa m}^3 \text{ mol}^{-1}$
Oxygen diffusivity	D_N	$1 \times 10^{-10} \text{ m}^2 \text{ s}^{-1}$

3.2 Contribution of carbon particle overlap to oxygen transport resistance

The local transport process of oxygen in ionomer is very complex. On the one hand, Pt particles are dispersed on the carbon surface, which will cause the transport path of oxygen to be longer than the actual ionomer thickness [15]. On the other hand, when the carbon particle overlap intensifies, the ionomer area may decrease significantly, resulting in insufficient oxygen supply. Both of these factors are considered in the modeling process, as shown in Eq. (30),

$$\delta_N^{\text{eff}} = \delta_N \times S_N / S_{Pt} \chi \quad (30)$$

where S is the surface area and χ denotes the oxygen supply coefficient. Assuming that Pt particles are evenly distributed on the carbon surface, the area ratio of carbon to ionomer can measure the supply of oxygen, so χ is defined as S_c / S_N . Although simple, this expression is effective to reflect the relationship between oxygen supply and ionomer surface area.

In this section, the case where I/C is equal to 0.95 is studied. δ is fixed to 3 nm, and k varies from 0 to 4. Different δ_N^{eff} is obtained and is presented in Fig. 3. Firstly, the Pt loading has a significant effect on the oxygen transport path. When k is 0, as Pt loading reduces from 0.4 to 0.02 mg cm^{-2} , δ_N^{eff} is increased from 4.59 nm to 91.80 nm. Note that k equal to 0 corresponds to no carbon particle overlap, and the increase of δ_N^{eff} is only caused by the increase of S_N / S_{Pt} (i.e., decrease of Pt loading). Secondly, the effect of carbon particle overlap is more significant under a low Pt loading. $\Delta\delta$ is defined as the δ_N^{eff} difference when k reduces from 4 to 0. It can be seen that as the Pt loading reduces from 0.4 to 0.02 mg cm^{-2} , the $\Delta\delta$ increases from 2.29 nm to 45.9 nm. In summary, both Pt loading and carbon particle overlap have significant effects on oxygen transport path in ionomer.

Previous studies [5, 9, 11] have found that there is a linear relationship between the local transport resistance and the reciprocal of roughness factor (f_{Pt} , the surface area of the catalyst per unit MEA area). According to the study of Greszler et al. [5], the slope of the line indicates the oxygen transport resistance on Pt surface R_{Pt} . From the micro morphology of the catalyst layer, R_{Pt} should be caused by the diffusion resistance of oxygen in ionomer. Many experimental studies also have found very large R_{Pt} . The measurement of Sakai et al. [16] is as high as nearly 1987 s m^{-1} , and the measurements of Ono et al. [9] and Greszler et al. [5] are also around 1200 s m^{-1} . The result of Owejan et al. [10] is relatively lower, but it is still close to 750 s m^{-1} . In this study, R_{local} is calculated and plotted in Fig. 4. It can be seen that when k is equal to 3, the slope of the line is 640 s m^{-1} ; and when k is equal to 4, the slope of the line is 1580 s m^{-1} . Because the effects of carbon particle overlap on δ_N^{eff} are considered, the calculated R_{Pt} is on the same order of magnitude as the literature values. It

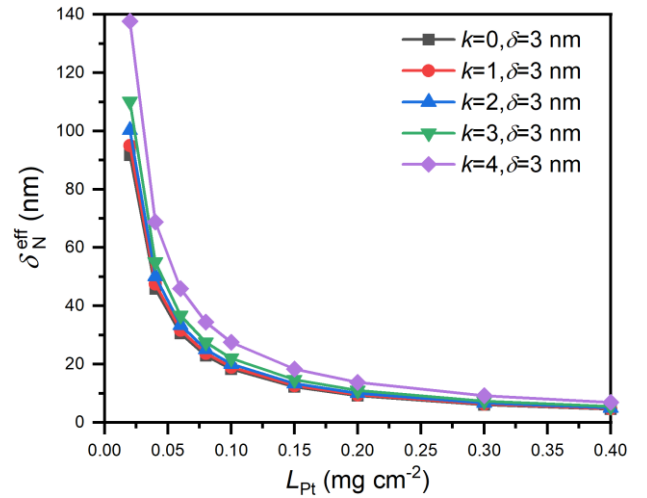


Fig. 3. Effects of L_{Pt} and k on δ_N^{eff} .

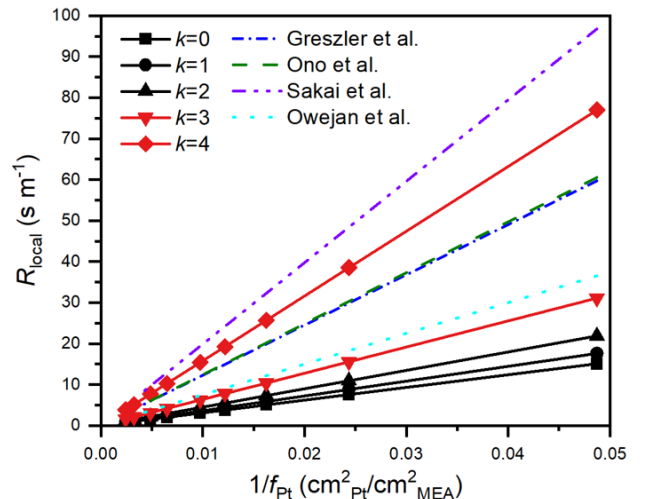


Fig. 4. Effects of $1/f_{Pt}$ and k on R_{local} .

shows that the increase of oxygen transport path can be successfully captured by our theoretical model, and the increased δ_N^{eff} as high as 140 nm under a low Pt loading can be considered as one of the sources of the Pt surface resistance R_{Pt} .

4. CONCLUSIONS

Carbon particle overlap in the catalyst layer will affect the morphology of the ionomer, and considering the influence of carbon particle overlap can provide a new insight into the origin of oxygen transport resistance. In this study, a theoretical catalyst layer model considering carbon particle overlap is constructed, and the effects of the overlap number and overlap degree on ionomer thickness are studied. It is found that the overlap of carbon particles will increase the physical thickness of ionomer and the effective transport path of oxygen in ionomer film. The effects of Pt loading on oxygen transport resistance are also studied. When the I/C is 0.95, with the decrease of Pt loading from 0.4 to 0.02 mg cm⁻², the effective transport path of oxygen even exceeds 100 nm, and the oxygen transport resistance under low Pt loading is successfully predicted. This also shows that the interfacial resistance may not be the only source of low-Pt-loading oxygen transport resistance, and the influence of carbon particle overlap on Pt/C particle structure can also be another possible reason.

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