論 文 の 内 容 の 要 旨

論文題目　A Polymer Electrolyte Fuel Cell Modeling Considering Microscopic Phenomena and Catalyst Activity

（ミクロな現 象及び触媒活性を考慮した固体高分子形燃料電池モデル）

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Recently, modeling approaches have become more and more important to the development of polymer electrolyte fuel cell (PEFCs) because they are less time-consuming, inexpensive, and, more versatile as compared with experimental approaches. In previous research, most models developed for PEFCs have been concentrated on heat and water transports in diffusion layer, gas flow channel, and electrolyte membrane. Unfortunately, electrochemical reaction occurring inside the catalyst layers has not yet been focused in detail. The lack of consideration of microscopic reaction phenomena in the catalyst layers makes PEFC models unrealistic, and causes various problems, for example, overestimation of cell performance in the reaction-dominated region, the use of fitting parameters instead of experimental parameters. Literature review of PEFC modeling was thoroughly presented in Chapter 1.

To overcome the reported problems and obtain a more accurate and realistic model, in this work, microscopic reaction phenomenon of adsorption of oxygen-containing species was considered and connected to macroscopic PEFC modeling. Firstly, in Chapter 2, experiments were carried out to determine pore level (either primary or secondary pore level), at which the rate-determining step of overall oxygen reduction reaction (ORR) takes place. It was found that diffusions and reactions in primary pores are negligible as compared with those in secondary pores due to the low penetration of Nafion ionomer into primary pores. This finding verified the validity of the cylindrical secondary pore-based modeling, which was further used as the macroscopic modeling for catalyst layers of PEFCs. The developed model can predict cell performance accurately in the low-potential region, however, without consideration of microscopic reaction phenomena, it overestimated cell performance in the high-potential region (reaction-dominated region).

In Chapter 3, the effect of microscopic phenomenon of adsorbed oxygen-containing species on

cell performance and Tafel slope were systematically investigated in practical, gas-phase PEFCs. The adsorbed oxygen-containing species are believed to be the main reason of the high activation overpotential, resulting in rapid drop of cell performance in the high-potential region. To introduce this poisoning effect into PEFC modeling, the quantity of the adsorbed oxygen-containing species was measured by using stripping voltammetry technique and it was further modeled as a reaction parameter so-called coverage factor as a function of potential. The coverage factor was taken into account in a modified Tafel reaction equation. With this improvement, the modified Tafel reaction equation can respond to the change in activation overpotential, and thus prediction of cell performance in the high-potential region was significantly improved.

In Chapter 4, the knowledge of adsorbed oxygen-containing species obtained from Chapter 3 was employed to analyze the effect of Pt particle size on ORR activity of Pt/C catalysts in practical, gas-phase PEFCs. Dependence of adsorbed oxygen-containing species on potential and relative humidity was almost identical for Pt sizes of 2 – 8 nm, which implied that there was an insignificant change in electronic properties of the Pt surface due to the variation of Pt particle size. As a result, no Pt particle-size effect on ORR activity was observed at any cell temperature ranging from 25 – 80 °C. Thus, it can be concluded that the ORR activity of a particular Pt/C catalyst was always the same, regardless of the Pt particle size, not only in a liquid-phase system but also in the a gas-phase system of practical PEFC MEAs. In addition, based on this result, my model can also be applied to predict cell performance of PEFC MEAs prepared with different Pt particle sizes.

In Chapter 5, a novel PEFC model, which focus on reaction parameters from microscopic to macroscopic aspects, was successfully developed. The modified Tafel equation taking microscopic poisoning effect of adsorbed oxygen-containing species developed in Chapter 3 was connected to the macroscopic cylindrical secondary pore-based model in Chapter 2 to complete a single-cell PEFC modeling. With this connection, the developed model can predict cell performance accurately in the entire potential range. This means the problems of overestimation and the use of fitting parameters were overcome, and thus the model can be more realistic. Subsequently, the developed model was used to demonstrate the importance of the poisoning effect, which should be solved in order to achieve a high-performance PEFCs operating at high potentials.

Finally, a summary of key findings in this work was presented, and the potential applications and the guide for future development of PEFCs discovered in this work were proposed in Chapter 6.