### Title

Modeling the Environment-Dependent Kinetics of Oxygen Reduction Reaction — a Continuum Model for Electric Double Layer




### Journal

ECS Meeting Abstracts, MA2023-02(37)

### ISSN

2151-2043



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### Publication Date

2023-12-22

### DOI

10.1149/ma2023-02371714mtgabs

Peer reviewed

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## Modeling the Environment-Dependent Kinetics of Oxygen Reduction Reaction – a Continuum Model for Electric Double Layer –

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**Abstract Text:**
For proton-exchange-membrane fuel cells (PEMFCs) to achieve broad commercialization, improved energy-conversion efficiency with minimal Pt-based electrocatalyst is required [1]. Because the sluggish rate of oxygen reduction reaction (ORR) limits the efficiency of PEMFCs [2], the efficiency improvement requires a better understanding of ORR kinetics and mechanism to design better catalyst. To understand the ORR mechanism, theoretical and experimental analyses have been conducted. While previous studies reasonably explained the catalyst-dependent activity on single crystal catalysts in 0.1 M perchloric acid solution [3, 4], the explicit effect of electrolyte and related microenvironments [5, 6] is not thoroughly understood. The change in the electrolyte alters the electric-double-layer (EDL) structure and thus the local microenvironment at the electrode/electrolyte interface. Thus, the structure of the EDL should be carefully analyzed to uncover the electrolyte-dependent reaction kinetics. In this talk, we propose a multiscale continuum model to predict the EDL structure and examine the effect of perchloric acid concentration on ORR activity on Pt (111). The model includes Density Potential Functional Theory (DPFT) for electron density and Modified Poisson Boltzmann equation for species' density and electric potential [7]. Also, the interaction between adsorbents and electric field is taken into account by minimizing the grand potential. After model validation with experimentally measured double-layer capacity data as a function of applied potential and concentration, the effect of the perchloric acid concentration (0.02 M – 0.2 M) on ORR activity is analyzed and discussed. It is shown that the model reproduces the specific activity obtained in the experiments when assuming the oxygen adsorption is limiting the rate, which can be attributed to the large energetic barrier for solvent reorganization [8]. Then, extension of the model to PEMFC ionomer electrolytes will be introduced. Overall, the model framework and findings provide insights into the ORR mechanism and guidance on how to tailor catalyst materials for increased PEMFC performance.

**Acknowledgements:**
This work was partially supported by the Hydrogen and Fuel Cell Technologies Office (HFTO), Office of Energy Efficiency and Renewable Energy, U.S. Department of Energy (DOE) through the Million Mile Fuel Cell Truck (M2FCT) consortia, technology managers G. Kleen and D. Papageorgopoulos, under contract number DE-AC02- 05CH11231, as well as by a CRADA with Toyota Central R&D Labs., Inc..

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**Symposium Selection:**
I01A - Polymer Electrolyte Fuel Cells and Electrolyzers 23 (PEFC&E23) - Diagnostics/Characterization Methods, MEA Design/ Model

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**Preferred Presentation Format:**
Oral

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