Energy Harvesting & Storage
Room Naupaka Salon 5-7 - Session EH-WeM2

Surfaces and Interfaces for Environmental Processes

Moderator: Craig Perkins, National Renewable Energy Laboratory

10:20am **EH-WeM2-8 Effect of Carbon Support Structures on Electrode Reaction Activity of Catalyst Layer in Polymer Electrolyte Fuel Cell: Large-scale Reactive Molecular Dynamics Simulations***, **Tetsuya Nakamura**, R. Otsuki, Y. Asano, Q. Chen, Y. Ootani, N. Ozawa, M. Kubo,* Tohoku University, Japan

For large output of polymer electrolyte fuel cell, the electrode reaction activity in the catalyst layer (CL) consisting of carbon support with meso pores, Pt nanoparticles (Pt NPs), Nafion chains, and water should be improved. Conventionally, to realize high electrode reaction activity, catalyst elements have been designed by first-principle calculation. However, first-principle calculation does not take into the meso-scale CL structures, such as carbon support, a distribution of the Nafion chains, and water content. Thus, we have constructed meso-scale CL structures and successfully clarified the effect of the pore size of the carbon support on the electrode reaction activity by large-scale reactive molecular dynamics simulation [1]. In addition, to design higher-performance CL structures, we consider it important to control the distribution of water on the carbon support and to improve the coverage states of Pt NPs with the Nafion chains. In this study, the hydrophilic group is introduced on the surface of carbon support to form a water film and we investigated coverage states of the Nafion chains over Pt NPs on the carbon support with different water film thicknesses.

The catalyst particle (CP) model was constructed by coating the Nafion chains and water on the carbon support surface. Six pores were created in an amorphous carbon sphere. In addition, the water film whose thickness is 0.7 and 1.0 nm were formed on the carbon support surface.

To clarify the effect of the thickness of water film on the coverage states of the Nafion chains over Pt NPs, we compared two CP models with different water film thicknesses of 0.7 and 1.0 nm. Here, for high electrode reaction activity, the Nafion chains should cover over Pt NPs partially to allow both high oxygen diffusion and high proton conduction. Figs. 2 (a) and (b) shows the coverage states of the Pt NPs interior of the pore after the calculations when the thickness of water film was 0.7 and 1.0 nm, respectively. Consequently, when the film thickness was 0.7 nm, the Nafion chains penetrated the pore and partially covered the Pt NPs. On the other hand, when the film thickness was 1.0 nm, the Nafion chains did not penetrate the pore. These results suggest that when the thickness of the water film is 0.7 nm, both high oxygen transport and high proton conduction to the Pt NPs in the interior of the pore of carbon support are realized.

[1] Tetsuya Nakamura, Riku Otsuki, Shuichi Uehara, Yuta Asano, Qian Chen, Yusuke Ootani, Nobuki Ozawa, Momoji Kubo, J. Comput. Chem. Jpn., 20, 4, 150-154 (2021)

10:40am **EH-WeM2-9 Large Scale Molecular Dynamics Simulation Study on Ionomer Coating of Pt Nanoparticles of Polymer Electrolyte Fuel Cells***, **Riku Otsuki**, T. Nakamura,* Institute for Materials Research, Tohoku University, Japan*; Q. Chen,* New Industry Creation Hatchery Center, Tohoku University, Japan*; Y. Asano, Y. Ootani,* Institute for Materials Research, Tohoku University, Japan*; N. Ozawa,* New Industry Creation Hatchery Center, Tohoku University, Japan*; M. Kubo,* Institute for Materials Research, Tohoku University, Japan

Polymer electrolyte fuel cells (PEFCs) are widely used for automobiles because it does not emit CO_2 during operation. For more popularization of a fuel cell vehicle, its higher output is required. The output of PEFC depends on a catalyst layer (CL) at the electrode composing of carbon supports, Pt nanoparticles (Pt NPs), ionomers and water molecules. Ionomers play an important role in proton conduction and oxygen gas diffusion. Especially, if Pt NPs and ionomer are not connected via water, proton transport is impossible. However, if the coverage of ionomer on Pt NPs is too high, oxygen gas diffusion is obstructed.Then, to reveal the CL structure with high electrode reaction activity, the coverage states of ionomer on Pt NPs have been earnestly studied by molecular dynamics (MD) method. For example, W. Wang et al. investigated the ionomer distribution over the Pt NPs and carbon particles in the CL model composed of about one hundred thousand atoms[1]. However, such a small model cannot handle the actual agglomerated structure of carbon support, and discussion about an optimal CL structure with high electrode reaction activity is impossible. In this study, we constructed 3 million atoms CL model with the agglomerated structure of the 15 carbon supports (Fig. 1) and analyzed the coverage of Nafion as ionomer over the Pt NPs by MD method to discuss the electrode reaction activity.

Coverage is defined as ratio of number of covered Pt atoms to the number of the Pt atoms on the Pt NPs surface. The distribution of Nafion coverage on each Pt NPs is shown in Fig. 2. The averaged coverage is 69.5%. Coverage of most Pt NPs, which placed on surface of a carbon support or between two carbon supports, ranges from 60 to 90%. On the other hand, it was found that the ionomer coverage of Pt NPs surrounded by three carbon supports is lower than another Pt NPs as shown in Fig. 3. Here, the Pt NPs and Nafion are connected via water clusters, indicating high oxygen gas diffusion and proton conduction. Therefore, we proposed that the Pt NPs surrounded by three carbon supports exhibit high electrode reaction activity. Finally, we conclude Large-scale MD is very effective to reveal how CL structure affect output characteristics of PEFCs

11:00am **EH-WeM2-10 Studying Corrosion Processes of Aluminum Alloys in Diverse Aqueous Environments***, **Micha Ben-Naim**, A. Ivanovskaya , S. Cho, C. Orme, M. Bagge-Hansen,* Lawrence Livermore National Laboratory

Corrosion modes and rates have a strong dependence on environmental conditions. During certain corrosion processes like stress corrosion cracking (SCC) and crevice corrosion, the local chemical environment, including pH and concentration of dissolved ions and gases, can vary dramatically from bulk solution conditions, motivating the study of corrosion properties under a wider range of aqueous environments than would typically be encountered in environmental corrosion. 7000 series aluminum alloys are a class of high-strength alloys used for structural and aerospace applicationsthat are susceptible to corrosion, particularly SCC, but work to date has largely focused on corrosion in aqueous chloride salts.

In this work, we study the corrosion properties of these Al alloys under a wide range of aqueous conditions. We explore new chemical environments that could lead to corrosion in Al alloys while probing the wide pH ranges that can arise in cracks and crevices. We use polished films to both measure bulk corrosion rates and enable the use of surface science techniques. Nondestructive electrochemical techniques enable high measurement throughput while measuring across pH and salt concentration ranges, so we measure corrosion potentials, corrosion rates, and electrochemical impedance spectra in a nondestructive manner. We also use surface characterization techniques (SEM, EDS, EBSD, XPS) to identify changes in surface chemistry and morphology. By combining electrochemical methods with surface science, we seek to gain a fundamental understanding of the corrosive regimes for these alloys.

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