Development of Advanced Electrocatalyst for Automotive Polymer Electrolyte Fuel Cells

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Platinum (Pt) or Pt-based electrocatalysts have been employed to facilitate electrochemical reactions such as hydrogen oxidation and oxygen reduction in polymer electrolyte fuel cells (PEFCs) because of their decent activity and stability comparing with other materials. However, due to Pt's high and fluctuating price, reduction of Pt loading on fuel cell electric vehicles (FCEVs) is strongly desired for FCEVs to be commercially viable. In order to reduce total Pt loading on FCEV, it is necessary to reduce total active area and/or Pt loading per unit active area without any sacrifice in performance of PEFC stack.

Increasing current density by improvement of mass transport through membrane electrode assembly (MEA) is effective to reduce total active area. It was demonstrated that introduction of thin polymer electrolyte membrane and fine flow channels has improved PEFC's performance especially at high current density range resulting in substantial reduction of Pt loading,[1] although further reduction is needed. Reduction of Pt loading per unit active area increases mass transport loss in catalyst layer (CL) as well as kinetic loss.[2] Hence, it is important to identify factors affecting transport properties in catalyst layer so that mass transport loss in CL would be minimized.

Activity enhancement for the sluggish oxygen reduction reaction (ORR) of Pt-based electrocatalyst is the most essential to reduce Pt loading. Although ORR activity improvement of classical homogeneous Pt-alloy nanoparticles has been leveling off at two- to four-fold compared to pure Pt nanoparticles, progress in the last decade in finely-tailored Pt-based electrocatalysts like core-shells, segregated Pt-alloys, nanostructured thin-film is remarkable.[3-6] In addition, our internally prepared Pt-based compound showed noticeable improvement in ORR activity though it is at the test-tube stage (Fig. 1).

Nissan, as an automotive manufacturer, has been devoting its energies to constructing evaluation techniques that can judge if a newly-developed electrocatalyst has a potential to be a candidate for FCEV application. Keeping in mind the automotive application, measurement of the activity over the entire operating range is desirable. Even though pure oxygen is used for cathode feed when *iV* performance of MEA is measured, influence of proton migration and oxygen diffusion within CL is superimposed on kinetic currents especially at high current density regime. A homogeneous 1-D CL model based on transmission line enables us to estimate catalytic activity in wide operating range.[7]

MEA testing is utilized to evaluate performance (activity, durability and robustness) of electrocatalyst under various operating conditions. Nevertheless, it is usually time-consuming and needs certain amount of electrocatalyst for MEA preparation. Employment of rotating disk electrode (RDE) technique is reasonable at an early stage of electrocatalyst development. Then, we have constructed an RDE evaluation scheme that covers various operating conditions so that applicability of developed electrocatalyst for FCEV can be estimated efficiently.

An understanding of performance-determining factors is required for not only rational design of advanced electrocatalyst but also controlling quality and reliability. Evolution of analysis and characterization techniques has greatly contributed to understanding critical parameters that affect performance of electrocatalysts. Especially, investigations using well-defined single-crystalline electrodes have provided many insights into relationship between surface structure and electrochemical characteristics.[8-10] However, there is still a lack of understanding of nanoparticle design that ensure every required performance, i.e. activity, durability and robustness. Accordingly, further evolution of fundamental analysis technology including theoretical calculations is expected as well as progress of electrocatalyst development.
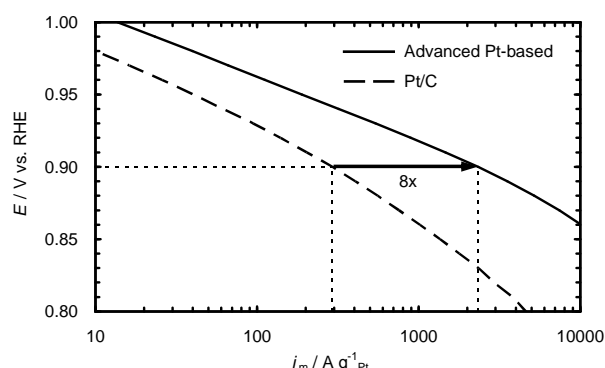

Fig. 1 Mass-transport and iR corrected Tafel plots of ORR obtained from RDE measurements in O_2-saturated 0.1 M $HClO_4$ at 25°C with anodic scan at 10 mV s^{-1}.

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